PROCEEDINGS
OF THE
NATIONAL ACADEMY OF SCIENCES
Volume 5 DECEMBER 22, 1919 Number 12

THE CHANGE OF MOLECULAR KINETIC ENERGY INTO MOLECULAR POTENTIAL ENERGY: THE ENTROPY PRINCIPLE AND MOLECULAR ASSOCIATION

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Communicated by J. Stieglitz, September 16, 1919

My work on the orientation of molecules in the surfaces of liquids (Langmuir has also worked on orientation) has led to the recognition of a remarkable new principle or law concerning the change of molecular kinetic energy into molecular potential energy. None of the kinetic relations already found in the very simple case where gases alone are involved, have been found to be as exact as the laws of thermodynamics, since by their very nature such relations are affected by many extraneous complicating factors. It need not then be surprising if similar relations involving the much more complicated and hitherto obscure kinetic phenomena of the liquid and the solid states should prove to be approximate rather than exact. Thus it is well known that Raoult's law concerning the kinetics of the vaporization of the various components in a solution, is exact only in an extremely limited range, that is when the components are practically alike with respect to cohesion[1] or with respect to the electromagnetic fields surrounding their molecules.[2] It is, therefore, somewhat startling to find that my new relation or law is, in one of its forms, much more generally applicable than Raoult's law, at least if the data at present accepted in this connection are as exact as they are supposed to be.

The new principle or law will first be stated in one of its special forms as follows: *Whenever a molecule moves from the interior of a liquid into the surface* in such a way as to form a new surface, *the average amount of its*

kinetic energy which is converted into potential energy is equal to 144% of the mean translational kinetic energy of a gas molecule at the same temperature. This indicates that in general only the faster moving molecules possess sufficient kinetic energy to carry them into the surface. All known plane surfaces have a positive free surface energy, that is the molecular potential energy in a plane surface in which the above principle holds is always greater than 144% of the mean kinetic energy of its molecules.

Application to the theory of surfaces.—As has been indicated in the last paragraph the molecules in a surface possess potential energy by virtue of their position. When a new surface is formed the principle given above indicates that a definite portion of this potential energy results from a tranformation of the kinetic energy of molecular motion into the potential form, and that the amount of energy supplied in this form depends only upon the temperature; and is proportional to the temperature. *The free energy of the surface is simply the difference between the total energy, which depends upon the structure of the surface, and the latent heat of the surface which is conditioned by the above law.* It is easy to see why, on this basis, the surface tension or the free surface energy decreases with the temperature. The total surface energy is approximately constant while the temperature is varied, provided the critical temperature is not too closely approached, which is the condition also for the application of the new principle since the surface film thickens as the critical temperature is neared. Since the contribution of the kinetic energy of molecular motion to this total energy is proportional to the absolute temperature, the free surface energy must decrease with the temperature.

The entropy law.—According to Lunn[3] heat has two measurable aspects, energy and entropy. The first law of thermodynamics relates to heat changes in which energy is, but entropy is not, conserved. The second law considers heat changes in which energy in the form of heat alone is not, but entropy is, conserved. The principle discovered by me has been stated above in terms of energy but it may be much more simply stated in terms of entropy as follows: *Whenever a molecule moves from the interior of a liquid into its surface in such a way as to form a part of a new surface, the entropy of surface formation is not only independent of the nature of the molecule, but is also independent of the temperature.* The numerical value of this entropy is 2.96×10^{-16} ergs per degree per molecule.

This entropy is not the ordinary thermodynamic entropy but is about

15% less than the thermodynamic entropy calculated for the area occupied by one molecule. The entropy which corresponds to the value given above is that which is calculated for the area of surface occupied by one molecule on the supposition that the arrangement of the molecules is such as to give approximately the same number of molecules in the surface as if it were built up on the plan of a cubic lattice, and the area for which the entropy is calculated is always that occupied by one molecule no matter what the temperature may be. The difference between these two entropies will be discussed later, but it is not essential *since either of them shows the constancy required by the relation under discussion.* When the extent of the molecular orientation in the surface is known it should be taken into account.

Relation of the entropy law to molecular association.—Although the entropy principle presented in this paper was discovered by the writer not more than a few years ago, a number of empirical relations which involve its validity without its recognition by their discoverers, have been well known for many years. The empirical relation directly related to the form of the entropy principle already given, is known as the law of Eötvös,[4] Ramsay, and Shields.[5] While a part of this relation was developed by Eötvös from a special form of the theorem of corresponding states, in its final form, as given by Ramsay and Shields, it may be considered as purely empirical, and it is this latter form which is given by the entropy principle. Indeed, though it does not seem to have been recognized by them, Ramsay and Shields relation gives the entropy of a surface, though in very peculiar units, and also the entropy expressed by it is not the ordinary thermodynamic entropy.

The thermodynamic entropy may be expressed in ergs per degree per square centimeter, or in other units. The Ramsay and Shields relation may be expressed in terms of entropy, if a somewhat unusual unit is used for the measurement of the area, in the following terms: *The entropy of the surface of a liquid is 2.12 ergs per degree for an area which is equal to the area of one face of a cube which contains one gram molecule of the liquid.* Since the volume of such a cube varies with the temperature, the area to which the entropy is referred also varies with the temperature.

The value in the use of such a peculiar system does not lie in the use of the same number of molecules in the box in every case, but in the use of the area of one face of the box, the inherent idea being that in this way the same number of molecules in the surface are obtained in every case when the molecules in the surface are counted by considering that their complexity in the surface to be the same as that inside the liquid.

It is obvious that this depends either upon the approximation to a cubic lattice arrangement, or to a similarity in the surface arrangement in every case. It is therefore to be expected that the relation would not hold in any case where the molecular orientation in the surface is such as to cause a deviation from such similarity.

While the normal value for the fraction of the molecular kinetic energy which is converted into the potential form is 144% of the mean molecular kinetic energy, which corresponds to a surface entropy of 2.96×10^{-16} ergs per molecule, and while what have been usually termed normal or unassociated liquids give these values very closely, certain liquids have been found for which the entropy value is lower. Thus the alcohols, organic acids, and water, give at ordinary temperatures not far from one-half the normal values (one-third in the case of water), but as the temperature increases, even for such liquids the entropy increases and approaches the normal value. Such results as these are easily explained on the basis of the assumption made by Ramsay and Shields and their followers in the association school, who consider that at low temperatures the molecules in such liquids are associated into larger groups than correspond to the formula weight, but that the association decreases as the temperature increases.

Criticisms of the Ramsay-Shields method of determining molecular association.—Certain sweeping criticisms of the Ramsay-Shields method of calculating the degree of molecular association which have been made, I consider to have no validity. On the other hand objections might be raised which seem not to have occurred to the workers in this field. The most sweeping criticism of the method, which has been expressed many times, is that since it is a "surface tension method it gives no true indication of the molecular state of the liquid as a whole." From this view-point it is considered that it is not the association in the liquid, but the association in the surface, which is calculated by this method. With this criticism I disagree most strongly, for if Ramsay and Shields have calculated any association at all it is not that in the surface, but that which exists in the body of the liquid, as seems to me apparent from the entropy principle, for the kinetic energy which is converted into the potential energy of the surface is the molecular kinetic energy of the molecules just before they move into the surface.

A second criticism which has been made is that very complex molecules, particularly those which contain several, paraffin chains such as tripalmatin and tristearin, give much too large values of the Ramsay-Shields constant. I have found, however, that a very great reduction

in the value of the constant is obtained if the molecular orientation in the surface is taken into consideration, which was not done by Ramsay and Shields. While the values which are thus obtained are still somewhat larger than the normal, it must be remembered that such complex molecules may vibrate in parts, which might very well give larger amounts of kinetic energy available for transformation into potential energy.

While Ramsay and Shields evidently did not see at all the important theoretical basis for their method it is nevertheless true that their equation involved the entropy of formation of the surface, and therefore kept closer to this basis than the very great number of supposedly improved equations which have been developed from it.

In so far as the experimental results for the temperature coefficients of free surface energy as determined by Ramsay and Shields can be trusted, our calculations show that the normal value of the surface entropy per degree per molecule is very close to the normal value (2.96×10^{-16} ergs) for the following substances over a wide temperature range: benzene, carbon tetrachloride, ethyl acetate, ethyl ether, methyl formate, and chlorobenzene. The results of Baly and Donnan give 2.8×10^{-16} for liquid nitrogen, and 2.75×10^{-16} for liquid oxygen.

On the other hand the values for argon (1.6×10^{-16}) and for mercury (1.5×10^{-16}), though not so well established experimentally, are not very much above 50% of the normal values, and the work of Jaeger indicates that molten salts also give low values. Unfortunately the data for argon were obtained over such a small temperature range that the entropy obtained may be very much in error. In the case of mercury and the molten salts, those who belong to the extreme association school would assume that the entire deviation is due to molecular association, but I am not at all convinced that the association is at all definite, especially since our present knowledge of the structure of metals and salts indicates that there may not be such a thing as a definite molecular weight in any case. This subject will be treated more comprehensively in a later paper.

A general form of the entropy principle.—In the preceding paragraph I have presented the entropy principle in its relation to the formation of surfaces, in which case it seems to hold remarkably well. Possibly it is only in the formation of surfaces that such a relation will be found to hold with exactness, as the following reasoning will indicate. In the formation of a surface the *total* energy converted into molecular potential energy must be of such a magnitude as to overcome the

cohesive forces to the extent which is necessary in the formation of the surface. However, as has already been stated, all of this energy does not come from the molecular motion, but a part of it is introduced by the action of an external force, which does the work necessary to produce what is called the free energy of the surface. The energy which is not supplied by the molecular motion, must be supplied in the form of work, so the amount of energy supplied by the molecular motion is not uniquely determined by the cohesive forces. When a liquid vaporizes in an ordinary experiment, the *whole* of the energy necessary for the separation of the molecules against the cohesive forces, is supplied by the energy of molecular motion, except for the amount which may be supplied by changes in the potential energy of the molecules themselves, and the latter is probably not a very important factor. It is obvious that the contribution of the molecular motion to the formation of a surface may be largely independent of the cohesive forces involved, and this is made probable by the validity of the entropy principle in this case. The cohesion enters so directly into the heat of vaporization that it would seem doubtful if for it the entropy principle would hold. It would seem surprising, too, if the principle should hold for melting, for sublimation, or for sublimation combined with dissociation.

Nevertheless, a study of the literature shows that while the entropy principle itself has hitherto remained unrecognized, there are a number of empirical relations, which if they hold, involve the validity of a much more general entropy principle. Without discussing to what extent I believe such a principle is valid, I will state it in two different forms as the theoretical basis of all of these empirical relations. In order to give this statement I will define a *region* as a phase, surface, or interface. *When in a system consisting of one component, a molecule moves from one region into another, the average molecular kinetic energy which is converted into the form of molecular potential energy depends in general only on the change of state, that is on the region from which the molecule comes and the one into which it goes, provided* that when a vapor phase is involved one condition which enters is the state of the vapor with reference to one variable. According to Trouton this condition is that the pressure of the vapor shall be the same in all cases, and this has been modified by Hildebrand, with an increase in accuracy, to the condition that the molecular concentration of the vapor phase shall be the same in all cases. In the second form, the principle states that the *entropy* of the change depends only on the change of state, and not on the individual nature of the molecules. In the application of this

principle the surface of a metal is not considered as a region of the same class as the surface of water or an organic liquid, since the characteristics of the two regions are quite unlike.

Corresponding to the various empirical relations the following 'normal' values of the entropy may be given:

	Entropy in ergs per degree per molecule $\times 10^{16}$
1. Liquid to surface	2.96
2. Liquid to vapor at the special concentration of:	
c = 0.00507 mols per liter	18.8
c = 0.0127 mols per liter	16.7
c = 0.0201 mols per liter	15.7
(These values become less accurate as the concentration of the vapor increases.)	
3. Solid to vapor at the melting point	21.0
4. Solid to liquid	9.0
5. Solids dissociate to 760 mm. vapor pressure	22.0

Of these the first is the most exact, the second holds moderately well under the conditions imposed, and the fourth, as might be expected, is one of the least accurate. Walden's rule is that the molar heat of fusion divided by the temperature is equal to 13.5 calories per degree for normal substances, or the molar entropy of fusion has the given value. This rule Walden[7] found to hold for a large number of organic substances. When the data did not correspond with what should be obtained according to the rule, Walden assumed that it still remains valid, but that the molecular weight is different from that given by the formula. However it is evident that this explanation is not sufficient to account for all of the deviations which exist.

At my request Mr. L. E. Roberts has studied practically all of the available data on the entropy of melting, and has found the general relations which hold. One of the greatest obstacles in this connection is that the data are in many cases extremely inaccurate. They indicate that the *latent heat of melting* of a metallic element which crystallizes in the regular system, *increases as the melting point rises*, and the entropy averages about 2.2 calories per gram atom per degree.[8] The salts show a somewhat similar relation, and at the same time there seems to be a general increase in the entropy of fusion with the number of gram atoms in the formula weight of the salt. There is a great deal of irregularity, but for the halogen salts the entropy is of the order of 2.2 calories per degree per gram atom, or about the same value as is found for the metals.[9] Hydrogen, hydroxides and water of crystallization are represented by lower values. Thus the entropy of fusion of the hydroxides of sodium, potassium, rubidium, and caesium, is about 2.8 calories per

degree per gram molecule. On turning to molecular compounds such as ammonia and carbon dioxide it is found that the entropy of fusion is 9.3 for the former and 8.9 for the latter, while for benzene the value is 8.3. Corresponding to Walden's rule a large number of organic compounds have entropies of fusion between 12 and 14 calories per degree, while many of the substances with smaller entropies of fusion possess other properties characteristic of associated liquids when they are in the liquid state. As might be expected substances with very complex formulae give high values, the increase with molecular complexity being very distinct. Thus with stearic acid ($C_1H_{36}O_2$) the value rises to 40, while the acid with two carbon atoms has an entropy of only 9.5, with nine carbon atoms of 10.5, while in the case of the 12 carbon atom acid the value rises to 27.

It is thus to be seen that the entropy is a very important function in a study of the transfer of molecules from one region to another, and that *in general the price which a molecule has to pay in terms of energy in order to undergo any certain change, increases with the temperature* or the molecules pay in proportion to their wealth with respect to energy. There is also an increase in this energy price whenever the complexity of the molecules increases sufficiently. The price in terms of entropy is much more constant than the price in terms of energy, and in this sense there is an analogy to the action of a Carnot engine.

It is believed that the point of view presented in abstract in this short paper will greatly change the present attitude in regard to the determination of molecular association in liquids, and it is possible that it may be of importance in a study of the general subject of the partition of energy, as well as in changes of kinetic into potential energy.[10]

The complete paper of which this is a part will be presented to the *Journal of the American Chemical Society* by Mr. L. E. Roberts and the writer, for publication in a later issue.

[1] *J. Amer. Chem. Soc., Easton, Pa.*, 38, 1916, (1452).

[2] *Ibid.*, 41, 1919, (970–92). PROCEEDINGS, May, 1919.

[3] *Physic. Rev.*, July, 1919.

[4] *Leipsig, Ann. Physik*, 27, 1886, (452).

[5] *London, Phil. Trans. Roy. Soc.*, 184A, 1893, (647), *Zs. Physik. Chem., Leipsig*, 12, 1893, (647).

[6] *J. Amer. Chem. Soc.*, 37, 1915, (975).

[7] *Zs. Elektrochem.*, 14, 1908, (715).

[8] Crompton, *J. Chem. Soc.*, 67, 1895, (315–327). T. W. Richards, *J. Franklin Institute*, 1902.

[9] Wayling, *Phil. Mag.*, 37, 1919, (495).

[10] Since this paper was submitted I have received a copy of a paper presented to the Société Française de Physique by M. J. Duclaux on June 6, 1919. He believes that there are quantities of energy of magnitude 6.6×10^{-18} ergs.

THE DISPLACEMENT OF THE GRAVITATING NEEDLE IN ITS DEPENDENCE ON ATMOSPHERIC TEMPERATURES

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Communicated October 8, 1919

1. Introductory.—In *Science* (**50,** pp. 214, 279, 1919) I communicated some of the early results, showing that the deflection of the needle of a gravitation apparatus varies in marked degree with the temperature on the outside of the building. I have since carried these experiments on for another month and the evidence has become more definitely interpretable. The work was done in a semi-subterranean room, in which the thermostat shows temperature variations which do not usually exceed a fraction of a degree. The room is large and so damp that all electrical excitation is excluded. Tests with radium fully confirmed this. Moreover the room is kept dark. The apparatus (Proceedings, **4,** p. 338, 1918) placed on the north-south wall of the pier confronts an eastern 30-inch wall, at a distance of about 4 meters and the outside of this is illuminated by sunlight, if present, in the morning, only.

2. Observations.—The observations during July and August are given at the bottom of the figure, the two curves being mean results of the a.m. and p.m. readings, respectively. The telescopic reading of the scale is y, so that Δy denotes the mean (static) excursion or double amplitude, when the attracting mass, $M = 1$ kgm. is passed from one side to the other of the attracted shot ($m = 0.6$ gram), at the end of a needle suspended by a quartz fiber. The actual excursion of the shot is $\Delta x = 0.01455\,\Delta y$, so that the magnification is about 70. The figure shows that even these mean excursions vary enormously, from values much below $\Delta y = 2$ to values above 7, easily five times. If individual excursions were taken, ratios as high as 10 might be found, in spite of the practically constant room temperature. On the upper part of the chart I have inserted the temperature observations θ in degrees F., made at Providence by the United States Weather Bureau, as well as the temperature variations $\Delta\theta$ (high minus low) of the successive days of the months, the same abscissas holding for all curves.

In the earlier data there seemed to be a close association between the Δy and θ curves. In the present data the regions $a, b, c, d, e,$ belong

together, though the Δy curve follows the θ curve with a lag of one or more days. A far better agreement in sense, not quantitatively always, now appears between the Δy and $\Delta\theta$ curves, and here in the given time scale, practically without a lag. To bring this to the eye more clearly, I have indicated the corresponding successive cusps in both curves with the same numbers 1 to 23. The agreement is in fact as close as it can possibly be, remembering that $\Delta\theta$ holds for twenty-four hours of the day and Δy only for the daylight interval of observation. In the same way the a.m. and p.m. curves differ enormously when there is sunlight, and very little in damp cloudy rainy weather (R in curve). In general

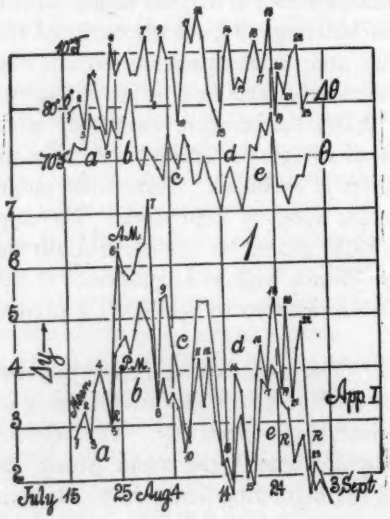

FIG. 1

and apart from details, the a.m. excursions reappeared in a subdued form in the p.m. results.

3. The needle in vacuum.—In *Science* I also communicated a series of results since much amplified, showing that for a case of two glass plates spaced by an impregnated wood frame, the initial attractions could be diminished to about one-third of their value by exhausting the case. The excursions diminished with the pressure, at a mean rate of 1%, per mercury centimeter of pressure. The glass plates in this case were about 1.8 cm. apart, inside. In case of the plenum the general character of the a.m. and p.m. excursion did not essentially differ from the graphs for apparatus I.

With the object of gaining some insight into the remarkable behavior at low pressure a new apparatus (no. III) was constructed with the glass plates spaced by a rectangular frame made of square brass tubing. The inside distance between plates was here 1.3 cm.; but in other respects it closely resembled the wood frame specified. The results with this metal case, however, differed totally from those of the other. In the morning of a bright day, there was usually marked repulsion between M and m, which changed gradually into an attraction at the close of the day. The repulsion was often so strong that the ends of the needle were pushed up into contact with the glass plates, to which position they returned whenever removed by tapping.

It was found, however, that the needle could be immediately freed by exhaustion of the case. In other words the repulsions passed continuously into attractions which were here at their maximum at the highest

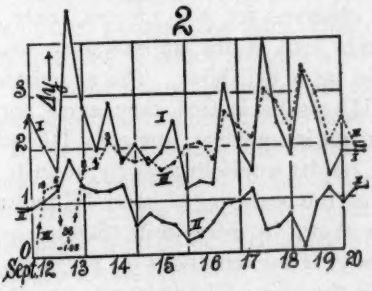

FIG. 2

exhaustions. The behavior of the metal case was thus the reverse of that of the wood case. In the former exhaustion removed a repulsion; in the latter, an attraction. It is difficult to assign a reason for this as there are three forces in contention: viz., gravitation and the radiant forces of the case (static) and of the external mass M. One is tempted to contrast the non-conducting wood with the conducting metal. The greater narrowness of the frame of the latter, however, gives the forces due to temperature distributions an advantage. In one respect the exhausted metal case has shown marked superiority; at a definite high vacuum, the excursions of the needle on any day are without drift; they are nevertheless variable on successive days. It is thus also improbable that this vacuum excursion corresponds to the gravitational attraction, so that an adequately trustworthy excursion is yet in arrears.

4. Record of the vacuum needle.—To exhibit these relations more clearly I have constructed figure 2, which contains a record of mean

results for three different apparatus, observations on the same vertical being made at the same time. The individual observations were taken thirty minutes apart Three or four means were deduced for the day. Apparatus I on the N.S. face of the pier fronting East has already been referred to in connection with figure I. Apparatus II was placed in a niche on the EW. wall of the pier fronting north, surrounding *on all sides* within 1 meter by the interior brick walls of the building. It thus receives secondary radiation only, and the graph in its details, departs utterly from curve I, particularly on clear days. If curves I and II were smoothed, however, they would show some resemblance.

Curve III are the results for the metal case (on the E.W. wall fronting south) with the needle kept in the partial vacuum, pressure p, marked on the curve. On the morning of September 12 and 13 at 39 and 36 cm. the two bodies M and m repelled each other. Even at lower pressures ($p = 8, 5, 3$, etc.) the results seemed fluctuating. Hence after September 14, I observed for $p < 1.5$ c.m., only (numeral omitted), there being a slight leak in the apparatus so that a higher vacuum could not be held for a half hour. The astonishing feature of these high vacuum (III) results is that they agree very closely with the observations (I) made in a plenum; whereas if III had also been observed in a plenum, the results would necessarily be in total opposition to I, as the repulsions at the beginning of curve III indicate.

Now it may be shown by direct tests (*Science*, l.c.) that the radiant forces of a hot body, M are repulsions for $p < 4$ cm. and attraction for higher pressures in the case. The exact pressure of radiant equilibrium resulting from this inversion is for incidental reasons difficult to specify; but one may estimate that in high vacua, y varies about 5 mm. per cm. of pressure p.

It follows from this that in the plenum apparatus (I) with eastern exposure, the attracting body M must be relatively warm in the morning and cold in the afternoon; while in the vacuum apparatus (III) with southern exposure, the body M is relatively cold in the morning and warm in the afternoon; for in such a case the radiant forces have the same sign. Hence the agreement in kind of the plenum graph, I, and the vacuum graph, III, in figure 2, after September 14, is a demonstration in a dark room, at practically constant temperature, of the rotation of the sun.

[1] Advance note from a Report to the Carnegie Institute of Washington, D. C.

CONDITIONS NECESSARY AND SUFFICIENT FOR THE EXISTENCE OF A STIELTJES INTEGRAL

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Communicated by E. H. Moore, October 8, 1919

The purpose of this note is to suggest a method for deriving a necessary and sufficient condition for the existence of the Stieltjes integral of $f(x)$ as to $u(x)$ from a to b in each of several forms generalizing those frequently employed (see *Encyclopédie des sciences mathématiques*, II_1, pp. 171–174) in the special case of Cauchy-Riemann integration—where $u(x)$ is of bounded variation and $f(x)$ is bounded on the interval (ab), $M \geqq f(x) \geqq m$. Bliss (Proceedings 3, 1917, pp. 633–637) has obtained one of these forms, perhaps the most satisfying of any; this note closes with his theorem, of which we give a new demonstration. Some of the other theorems stated (though here derived otherwise) are immediate consequences of the one due to Bliss or are otherwise intimately related to it, as the reader will readily see. Since it is believed that the present treatment will be found useful in connection with that of Bliss, our notation has been made to conform to his; moreover, reference to his paper is given for such isolated steps in the proof as may readily be supplied from it.

For a given partition π of (ab) due to the points $x_0 = a, x_1, x_2, \ldots, x_{n-1}, x_n = b, 0 < x_i - x_{i-1} < \delta$, form the sums

$$s_\pi = \sum_{i=1}^{n} M_i \Delta_i u, \quad S_\pi = \sum_{i=1}^{n} f(X_i) \Delta_i u, \quad \sigma_\pi = \sum_{i=1}^{n} m_i \Delta_i u,$$

where $\Delta_i u = u(x_i) - u(x_{i-1})$, X_i is any point of the interval (x_{i-1}, x_i), and $M_i[m_i]$ is the least upper bound [the greatest lower bound] of $f(x)$ on (x_{i-1}, x_i). If the limit of S_π exists as δ approaches zero, this limit is the Stieltjes integral of $f(x)$ as to $u(x)$ from a to b. In case $u(x)$ is a monotonic non-decreasing function, the limit, if existing, of $s_\pi[\sigma_\pi]$, as δ approaches zero, is here called the upper [lower] Stieltjes integral of $f(x)$ as to $u(x)$ from a to b.

Let us determine conditions under which the upper integral shall certainly exist when $u(x)$ is monotonic non-decreasing. If we form a repartition π' of (ab) as to π by taking the points forming π and certain additional points, it is clear that $S_{\pi'} \leqq S_\pi$. Moreover, there is ob-

viously a lower bound to $S_{\pi'}$. Hence if the number of divisions of (ab) is increased by repartitions in such wise that δ approaches zero, the sum $S_{\pi'}$ approaches a definite finite limit. Let us ask under what conditions we shall be led to a contradiction by supposing that two convergent sequences of sums σ_π for sequences of partitions π (whether formed by repartitions or not) with norms δ approaching 0 lead to different limits N_1 and N_2 where $N_1 < N_2$. Let ξ and η be two arbitrarily small positive numbers such that $N_1 + \xi + \eta < N_2$. Let π_1 be a partition of (ab) into s intervals belonging to the sequence by which N_1 is defined and let s be so great that $S_{\pi_1} < N_1 + \xi$. Let π_2 be a partition of (ab) into t intervals where t is an integer greater than s and subject to being made as large as one pleases. Let π_3 be a partition formed by the points of π_1 and π_2, so that π_3 is a repartition of both π_1 and π_2. Then we have

$$S_{\pi_2} - S_{\pi_3} \leqq \sum_{\alpha=1}^{s-1} (M_{i_\alpha} - m_{i_\alpha}) \Delta_{i_\alpha} u,$$

where the intervals $(x_{i_\alpha - 1}, x_{i_\alpha})$, obviously at most $s - 1$ in number, are all the intervals of π_2 which are separated into parts in forming π_3. Since t may be made large at our choice and since the sum in the second member of the foregoing relation never has more than $s - 1$ terms, whatever the value of t, it is clear that t may be chosen so large that this sum is less than η provided that $u(x)$ is continuous at the discontinuities of $f(x)$. Then we have $S_{\pi_3} \leqq S_{\pi_2} + \eta$. But we have seen that $S_{\pi_2} \leqq S_{\pi_1} < N_1 + \xi$. Hence we have $S_{\pi_3} < N_1 + \xi + \eta$. But, as t increases, S_{π_3} approaches N_2. Hence we have $N_2 \leqq N_1 + \xi + \eta$, contrary to the hypothesis $N_1 + \xi + \eta < N_2$. Hence the upper integral of $f(x)$ as to a monotonic non-decreasing function $u(x)$ exists provided that $u(x)$ is continuous at the points of discontinuity of $f(x)$. The corresponding result may likewise be proved for the case of a non-decreasing function $u(x)$; and also for the case of the lower integral. Hence, since every function of bounded variation $u(x)$ may be expressed as the difference of two monotonic non-decreasing functions which are continuous at the points of continuity of $u(x)$, we have the following theorem:

THEOREM I. *If $u(x)$ is of bounded variation and $f(x)$ is bounded on (ab) and if $f(x)$ is continuous at the points of discontinuity of $u(x)$, then the limits as δ approaches 0 of the sums s_π, σ_π of the preceding paragraph both exist.*

Let us now write the function $u(x)$ in the form

$$u(x) = u(a) + P(x) - N(x),$$

where $P(x)$ and $N(x)$ are respectively the positive and the negative variation of $u(x)$ on (ax); and by $U(x)$ denote the total variation $P(x) + N(x)$. Then $P(x)$, $N(x)$, $U(x)$ are continuous at the points of continuity of $u(x)$.

A sufficient condition for the existence of the integral of $f(x)$ as to $u(x)$ from a to b is the existence of the integral of $f(x)$ as to $P(x)$ and as to $N(x)$. Sufficient to this is the existence and equality of the upper and lower integrals of $f(x)$ as to $P(x)$ and as to $N(x)$. In case $u(x)$ is continuous or $f(x)$ is continuous at the discontinuities of $u(x)$, these upper and lower integrals surely exist and a sufficient condition for their equality is that the sums

$$\sum_{i=1}^{n}(M_i-m_i)\{P(x_i)-P(x_{i-1})\} \text{ and } \sum_{i=1}^{n}(M_i-m_i)\{N(x_i)-N(x_{i-1})\}$$

shall have the limit zero as δ approaches zero. Hence a sufficient condition for the existence of the integral of $f(x)$ as to $u(x)$ from a to b is that

$$\lim_{\delta=0}\sum_{i=1}^{n}(M_i-m_i)\{U(x_i)-U(x_{i-1})\}=0. \tag{1}$$

Bliss (l. c., p. 634, ll. 1–10) has shown that a necessary condition for the existence of the integral is that

$$\lim_{\delta=0}\sum_{i=1}^{n}(M_i-m_i)\,|\,u(x_i)-u(x_{i-1})\,|=0. \tag{2}$$

From this necessary condition if follows readily that $f(x)$ must be continuous at the points of discontinuity of $u(x)$ (see Bliss, l. c., p. 636, ll. 7–17). If we write $u(x) = v(x) + j(x)$ (Bliss, p. 636, ll. 1–7), where $j(x)$ is the function of 'jumps' of $u(x)$, it may be shown (Bliss, p. 636, ll. 18–36) that the integral of $f(x)$ as to $j(x)$ exists whenever that as to $u(x)$ exists. In the same way it may be shown that the integral of $f(x)$ as to $J(x)$ must also exist, where $J(x)$ is the total variation of $j(x)$ on the interval (ax). Hence a necessary condition for the existence of the integral of $f(x)$ as to $u(x)$ is the existence of the integral of $f(x)$ as to $v(x)$.

We propose to show next that the existence of the integral of $f(x)$ as to $u(x)$ implies that of $f(x)$ as to $U(x)$. In view of the results of the

preceding paragraph and of the fact that $U(x)$ is obviously equal to $V(x) + J(x)$, where $V(x)$ is the total variation of $v(x)$ on (ax), it is obviously sufficient to prove this for the case when $u(x)$ is a continuous function. Now, when $u(x)$ is continuous, we have (Vallée Poussin, *Cours d'Analyse*, vol. 1, 3rd edn, p. 73)

$$\lim_{\delta=0} \sum_{i=1}^{n} | u(x_i) - u(x_{i-1}) | = U(b), \tag{3}$$

with a like relation when b is replaced by x and the interval (ab) by the interval (ax). Since no $| u(x_i) - u(x_{i-1}) |$ is greater than the corresponding difference $U(x_i) - U(x_{i-1})$ and since the sum of the latter differences, for $i = 1, 2, \ldots, n$, is $U(b)$, we see that

$$\lim_{\delta=0} \sum_{i=1}^{n} [U(x_i) - U(x_{i-1}) - | u(x_i) - u(x_{i-1}) |] = 0 \tag{4}$$

and that no bracketed term here is negative. Hence for every ϵ there exists a δ_1 such that the i^{th} term ($i = 1, 2, \ldots, n$) of the sum in (4) is less than ϵ when $\delta < \delta_1$. Hence, for such δ, we have

$$0 \leqq \sum_{i=1}^{n} (M_i - m_i)[U(x_i) - U(x_{i-1}) - | u(x_i) - u(x_{i-1}) |] < (M - m)\epsilon \tag{5}$$

Hence, as δ approaches zero the sum in (5) approaches zero as a limit. This result and relation (2) imply (1). But the latter is sufficient to the existence of the integral of $f(x)$ as to $U(x)$, and indeed as to $u(x)$.

We are thus led to the following theorem:

THEOREM II. *If $u(x)$ is of bounded variation and $f(x)$ is bounded on (ab), then a necessary and sufficient condition for the existence of the integral of $f(x)$ as to $u(x)$ is the existence of the integral of $f(x)$ as to $U(x)$.*

Since (1) and (2) are identical when $u(x) = U(x)$ we now have readily the following theorems:

THEOREM III. *A necessary and sufficient condition for the existence of the integral of the bounded function $f(x)$ as to the function $u(x)$ of bounded variation is that the upper and lower integrals of $f(x)$ as to the total variation function $U(x)$ of $u(x)$ shall exist and be equal.*

THEOREM IV. *A necessary and sufficient condition for the existence of the integral from a to b of the bounded function $f(x)$ as to the function $u(x)$ of bounded variation is that the total oscillation of $f(x)$ as to $u(x)$ from a to b shall be zero, that is, that*

$$\lim_{\delta=0} \sum_{i=1}^{n} (M_i - m_i)[U(x_i) - U(x_{i-1})] = 0.$$

THEOREM V. *In order that a bounded function $f(x)$ shall be integrable from a to b as to a function $u(x)$ of bounded variation, it is necessary and sufficient that the interval (ab) may be divided into partial intervals so that the total variation of $u(x)$ in those in which the oscillation of $f(x)$ is greater than an arbitrarily preassigned positive number ω shall also be as small as one wishes.*

THEOREM VI. *If $u(x)$ is of bounded variation and $f(x)$ is bounded on the interval (ab), then a necessary and sufficient condition for the existence of the integral of $f(x)$ as to $u(x)$ from a to b is that the total variation of $u(x)$ on the set D of discontinuities of $f(x)$ shall be zero (Theorem of Bliss).*

Of the four preceding theorems the only one needing further proof is the last. [For the definition of the total variation of $u(x)$ on a set of points, see Bliss, l. c., p. 633, ll. 12–19.] Let $\epsilon_1, \epsilon_2, \epsilon_3, \ldots$ be a sequence of positive numbers decreasing monotonically toward zero, and let $D_1, D_2, D_3, \ldots$ be the closed set of points at which the oscillation of $f(x)$ is $\geqq \epsilon_1, \geqq \epsilon_2, \geqq \epsilon_3, \ldots$ Then the set D of discontinuities of $f(x)$ is the limit of the set D_n when n is indefinitely increased. Now if $f(x)$ is integrable as to $u(x)$ we have seen that the interval (ab) may be divided into partial intervals so that the total variation of $u(x)$ on those in which the oscillation of $f(x)$ is greater than an arbitrarily preassigned positive number shall be as small as one pleases; and this implies that the total variation of $u(x)$ on D_n, and hence on D, is zero. Again, if the total variation of $u(x)$ on D is zero so is it on D_n for every n; and hence $f(x)$ is integrable as to $u(x)$ since it is such that the interval (ab) may be divided into partial intervals so that the sum of those in which the oscillation is greater than an arbitrarily preassigned positive number shall be as small as one pleases.

TRANSFORMATIONS OF CYCLIC SYSTEMS OF CIRCLES

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Communicated by E. H. Moore, October 9, 1919

When two surfaces, S and $\bar{S}$, are applicable, there is a unique conjugate system on S which corresponds to a conjugate system on $\bar{S}$. Denote these conjugate systems, or *nets*, by N and $\bar{N}$ respectively.

The cartesian coördinates x, y, z, of N and $\bar{x}$, $\bar{y}$, $\bar{z}$ of $\overline{N}$ are solutions of an equation of the form

$$\frac{\partial^2\theta}{\partial u\partial v} = a\frac{\partial\theta}{\partial u} + b\frac{\partial\theta}{\partial v}, \tag{1}$$

which we call the common *point equation* of N and $\overline{N}$.

Let M and $\overline{M}$ be corresponding points of N and $\overline{N}$. With M as center describe a sphere whose radius is the distance from $\overline{M}$ to the origin. Let Σ_1 and Σ_2 be the sheets of the envelope of the spheres as M moves over N, and let μ_1 and μ_2 be the points of contact with Σ_1 and Σ_2 of the sphere with center at M. The null spheres with centers at μ_1 and μ_2 meet the tangent plane of N in a circle C. These ∞^2 circles from a cyclic system, that is they are orthogonal to ∞^1 surfaces.[1]

If h and l are any pair of solutions of the system

$$\frac{\partial h}{\partial v} = (l-h)\,a, \qquad \frac{\partial l}{\partial u} = (h-l)\,b, \tag{2}$$

the functions x', y', z', defined by the quadratures

$$\frac{\partial x'}{\partial u} = h\frac{\partial x}{\partial u}, \frac{\partial x'}{\partial v} = l\frac{\partial x}{\partial v}; \frac{\partial y'}{\partial u} = h\frac{\partial y}{\partial u}, \frac{\partial y'}{\partial v} = l\frac{\partial y}{\partial v}; \frac{\partial z'}{\partial u} = h\frac{\partial z}{\partial u}, \frac{\partial z'}{\partial v} = l\frac{\partial z}{\partial v}, \tag{3}$$

are the coördinates of a net N' parallel to N, and the functions $\bar{x}'$, $\bar{y}'$, $\bar{z}'$, defined by

$$\frac{\partial \bar{x}'}{\partial u} = h\frac{\partial \bar{x}}{\partial u}, \frac{\partial \bar{x}'}{\partial v} = l\frac{\partial \bar{x}}{\partial v}; \frac{\partial \bar{y}'}{\partial u} = h\frac{\partial \bar{y}}{\partial u}, \frac{\partial \bar{y}'}{\partial v} = l\frac{\partial \bar{y}}{\partial v}; \frac{\partial \bar{z}'}{\partial u} = h\frac{\partial \bar{z}}{\partial u}, \frac{\partial \bar{z}'}{\partial v} = l\frac{\partial \bar{z}}{\partial v}, \tag{4}$$

are the coördinates of a net $\overline{N}'$ parallel to $\overline{N}$. Moreover, the nets N^2 and $\overline{N}'$ are applicable, and consequently the function $\theta' = \Sigma x'^2 - \Sigma\bar{x}''$ is a solution of the point equation of N' and $\overline{N}'$. By the quadratures

$$\frac{\partial\theta}{\partial u} = \frac{1}{h}\frac{\partial\theta'}{\partial u}, \quad \frac{\partial\theta}{\partial v} = \frac{1}{l}\frac{\partial\theta'}{\partial v}$$

we obtain a solution θ of (1).

The functions x_1, y_1, z_1, and $\bar{x}_1$, $\bar{y}_1$, $\bar{z}_1$, defined by equations of the form

$$x_1 = x - \frac{\theta}{\theta'}x', \qquad \bar{x}_1 = \bar{x} - \frac{\theta}{\theta'}\bar{x}' \tag{5}$$

are the cartesian coördinates of two applicable nets N_1 and $\overline{N}_1$, which are T transforms of N and $\overline{N}_1$ respectively; a net and a T transform

are such that the developables of the congruence of lines joining corresponding points of the two nets meet the surfaces on which the nets lie in these nets.[2]

Since N_1 and $\overline{N}_1$ are applicable nets, we can obtain a cyclic system of circles C_1, in the manner described in the second paragraph. Hence each pair of solutions of (2) determines a transformation of the cyclic system of circles C into a cyclic system of circles C_1, such that the nets enveloped by the planes of the circles C and C_1 are in the relation of a transformation T. Moreover, it can be shown that corresponding circles lie on a sphere. We say that two such cyclic systems are in relation T.

Darboux[3] has stated the results of the second paragraph in the following form: If a surface $\bar{S}$ rolls over an applicable surface S, and Q is a point invariably fixed to S, the isotropic generators of the null sphere with center Q meet the plane of contact of S and $\bar{S}$ in points of a circle C which generate the surfaces orthogonal to the cyclic system of circles C. Making use of these ideas, we give the following interpretation of the above transformations of cyclic systems:

If N and $\overline{N}$ are applicable nets, and N_1 and $\overline{N}_1$ are respective T transforms by means of (5), where $\theta' = \Sigma x'^2 - \Sigma \bar{x}'^2$, the cyclic systems in which a point sphere invariably bound to $\overline{N}$ and $\overline{N}_1$ meets the planes of contact, as $\overline{N}$ rolls on N and $\overline{N}_1$ on N_1, are in relation T.

It can be shown that the two surfaces orthogonal to these respective cyclic systems which are generated by the points where an isotropic generator of the null sphere meets the plane of contact are in the relation of a transformation of Ribaucour, that is, these surfaces are the sheets of the envelope of a two parameter family of spheres and the lines of curvature on the two sheets correspond.

[1] Guichard, *Ann. Sci. Ec. norm., Paris*, (Ser. 3), 20, 1903, (202).
[2] Eisenhart, these PROCEEDINGS, 3, 1917, (637).
[3] *Leçons sur la théorie générale des surfaces*, vol. 4, 123.

NEW MEASUREMENTS OF THE VAPOR PRESSURE OF MERCURY

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Communicated by W. A. Noyes, October 14, 1919

Writing in 1908, Laby[1] remarked "The vapor pressure of mercury is intrinsically important; it has been determined for a wider range of temperatures than that of any other substance. . . . Yet the greatest—and, it should be added, unnecessary—disagreement is to be found in the current values for this vapor pressure, nor is there any table combining all the existing observations." Laby thereupon proceeded to collate the best observations available at that time and published a weighted average table.

In 1910 Smith and I published[2] the results of 43 direct observations of the vapor pressure of mercury by a static method over the temperature range 250° to 435°. The pressure measurement was immensely facilitated by the use of the newly-devised 'static isoteniscope;'[3] and a good form of platinum resistance thermometry was available. The average divergence in temperature of an individual observation from the smooth curve given by the equation

$$\log p = 9.9073436 - 3276.628/\theta - 0.6519904 \log \theta, \qquad (R)$$

whose constants were chosen to fit our results, was 0.050°. Because of the rather high degree of consistency thus attained, we were encouraged to exterpolate the curve in both directions and found that the values so obtained agreed remarkably well with the average experimental findings of those who had worked either above or below our temperature region.[4] A critical discussion of our own and of the older work on this subject, with tabular comparisons, may be found in the paper referred to.[2]

In 1909, Knudsen developed a relationship[5] connecting the weight of gas passing through tubes containing pierced diaphragms with the difference of gas pressure at the ends, the density of the gas, the resistance of the tube and diaphragm and the time of flow. He applied this to measure the vapor pressure of mercury[6] from 0° to 50°, above which temperature his relationship failed to hold. From the lower to the upper end of this range, his results run from 10.9 to 6.2% respectively in pressure lower than the values given by extrapolation of our 1910 results

by the above equation. This corresponds to temperature discrepancies of 1° at 0° and 0.8° at 50°. One regrets that such an elegant method is not more direct. Later, Knudsen[7] applied his admirable 'absolute manometer,' under certain difficulties, to measure this vapor pressure from −10° to +24.4°, and obtained results ranging from about 5% higher to about 10% lower in pressure, at the respective ends of this range, than the values furnished by our equation.

In 1913 Villiers[8] obtained a series of values from 60° to 100° which ran higher than ours by 2.7% of our calculated pressure at 60° to 7.5% at 100°.

In 1914, Haber and Kirschbaum,[9] adopting a suggestion of Langmuir's,[10] measured the damping of a quartz fiber vibrating in mercury vapor at 20°, and obtained a single value by an independent method. This value lies 3.8% in pressure below that given by our equation, or, otherwise, differs by 0.41° for the same pressure.

In glancing over the results obtained by these various workers, here stated baldly without criticism, one is inclined to believe that our equation of 1910 continues to stand the test of time by averaging their scattered results as well as can be hoped for. At the same time one is impressed by the pre-war activity in this field, especially in the lower temperature ranges. It appeared, therefore, to be of interest to obtain, by a method as direct as possible, new measurements below 250°, the lowest point of the range studied in 1910; and, in the work here described I have extended that range from 250° to 120°.

As the static isoteniscope is not especially well suited for the measurement of the rather low pressures with which we are here concerned, an entirely new plan was adopted. Two McLeod gauges of suitable capacities were calibrated and sealed to a central pressure reservoir. The gauges, which were operated by gas pressure and without rubber connexions, contained purified mercury. The reservoir and connected gauges could be charged with dry hydrogen at any pressure desired, and sealed off by mercury. The smaller of the two gauges was completely immersed in a rictously stirred oil bath whose temperature was measured by a mercurial thermometer whose thread was all submerged. The corrections of this thermometer were known[11] to tenths of a degree. The other, larger, gauge and the reservoir were maintained in baths at room temperature. On operating the two gauges simultaneously, different pressure readings were obtained, due to the condensation of the mercury vapor that contributed part of the total pressure in the hot gauge; and from this difference in reading the vapor pressure of mercury at the

temperature of the hot gauge could be computed in an obvious manner, if Dalton's law were assumed.

For details of the experiments and mode of reduction of the results, as well as for tabular comparison with the heretofore existing data, a paper may be consulted that will appear elsewhere.[12] It should, however, perhaps be stated here that, at the pressures and compressions used, the neglect of the known divergence of hydrogen from the simple gas laws would cause an error very much smaller than other errors of the experiment. Equlibrium in the hot gauge was, apparently, always established sooner than a measurement could be made. A small correction was applied to correct for the vapor pressure of mercury in the cold gauge. A possible systematic error may arise from the fact that the apparent vapor pressure of a liquid in contact with a gas is not in all cases identical with the vapor pressure at the same temperature of the same liquid in a vacuum. Although this fact was observed by Regnault,[13] it has apparently escaped the notice of many users of the gas-current method for determining small vapor pressures. The error is the greater the more soluble the gas in the liquid, although it is not conditioned solely by this solubility as such. With hydrogen gas at atmospheric pressure, the order of this error,[14] in the case of liquids in which hydrogen shows marked solubility such as ether, carbon disulphide and acetone, is about 1% at 30°; with mercury, in which no evidence of solution was found, and at the temperatures here in question, the order of the error might be expected to be much smaller, were it not that low gas pressures may be especially prejudicial. Further study of this possible error is planned, and one can only say, with T. W. Richards, that these results are of but a preliminary character.

The brief table below summarizes the results obtained. The 'calculated' values in the third column are those derived from the Kirchoff-Rankine-Dupré equation (R), whose constants were given above; the values in the fifth column are likewise derived from this equation. In the last column, P stands for the pressure, in millimeters of mercury, of hydrogen in the reservoir.

Vapor pressures of mercury

TEMPERATURE	VAPOR PRESSURE IN MILLIMETERS OF MERCURY AT 0°		$\frac{\text{FOUND}}{\text{CALCULATED}} \times 100$	PERCENTAGE CHANGE OF PRESSURE PER 0.1°	P
	Found	Calc.			
191.5°	13.02	13.02	100.00	0.34	14.28
150.0°	2.802	2.811	99.67	0.39	3.564
121.8°	0.829	0.823	100.75	0.47	1.738

Since the values thus found for the vapor pressures of mercury are obtained, broadly, by subtracting the reading of the smaller and less accurate hot gauge from those of the larger and more accurate cold gauge, it will be seen that the greatest precision (so far as gauge readings are concerned) is to be expected when the vapor pressure most closely[15] approaches P. This is borne out by the results as tabulated.

It is evident that the results found agree with those calculated almost as closely as can be expected, even having regard to the temperature error (0.1°) alone. For this reason one may state, meantime, that the equation (R) may be applied, without alteration of its constants, to give the vapor pressure of mercury over the range 120° to 250° as well as from 250° to 435°, as previously found. A table of the values given by this equation for every two degrees from 0° to 458° has already been published.[2] Further experimental extension of the temperature range, as well as greater precision, may be attempted later, should facilities be available, either by this or else by another method shortly to be described.

It is, perhaps, of interest to consider the upper temperature limit of this vapor pressure curve, namely the critical temperature of mercury. This has been the subject of much speculation.[16] Recently, in 1916 and 1917, J. J. van Laar, employing certain theoretical considerations, has computed 969°C.[17] and later 899°C.[18] for the critical temperature; while, in 1918, E. Ariès,[19] from other considerations, arrives at 1077°C. In such cases, a little experiment is worth much theory. At the Minneapolis meeting of the American Chemical Society in 1910, I described[20] some experiments in which I had heated mercury in a thick-walled capillary tube of quartz glass of 0.2 mm. bore, and continued to see, through a telescope, liquid and gas phases distinct up to 1275°C. At this temperature the meniscus disappeared; but this was found to be due, not to the attainment of the critical temperature, but to the slight blowing out of the quartz capillary with consequent enlargement of its volume. The tube was never ruptured, but the very viscous liquid quartz glass had yielded somewhat to relieve the rather high pressure of several hundred atmospheres. In 1912, Koenigsberger,[21] using a quartz capillary of dimensions very similar to mine, reported that he had reached the critical temperature about 1270°C; but I was able to point out[22] the cause of Koenigsberger's mistake. Owing to lack of facilities, I have not been able to develop this work further; but this has now been done, by the same capillary quartz tube and telescope method, by Bender[23] at Freiburg, who finds that the critical temperature lies between 1500° and 1600°C.

[1] Laby, *London, Phil. Mag.*, (Ser. 6), **16**, 1908, (789).

[2] Smith and Menzies, *J. Amer. Chem. Soc.*, **32**, 1910, (1434).

[3] Smith and Menzies, *Ibid.*, **32**, 1910, (1412).

[4] Pfaundler, *Leipsig, Ann. Physik*, (Ser. 3), **63**, 1897, (36); Morley, *Phil. Mag.*, (Ser. 6), **7**, 1904, (662); Hertz, *Ann. Physik.*, (Ser. 3), **17**, 1882, (193); Regnault, *Paris, Mém. Acad. Sci.*, **21**, 1847, (30, 502); **26**, 1862, (506); Ramsay and Young, *J. Chem. Soc.*, **49**, 1886, (37); Young, *Ibid.*, **59**, 1891, (629); Cailletet, Colardeau and Rivière, *Paris, C. R. Acad. Sci.*, **130**, 1900, (1585), etc.

[5] Knudsen, *Ann. Physik*, **28**, 1909, (1002).

[6] Knudsen, *Ibid.*, **29**, 1909, (184).

[7] Knudsen, *Ibid.*, **32**, 1910, (809).

[8] Villiers, *Ann. Chim. Phys.*, (Ser. 8), **30**, 1913, (588).

[9] Haber and Kirschbaum, *Zr. Elektrochemie.* **20**, 1914, (301).

[10] Langmuir, *J. Amer. Chem. Soc.*, **35**, 1913, (105).

[11] Reichsanstalt certificate dated 1914.

[12] Menzies, *J. Amer. Chem. Soc.*, probably November, 1919.

[13] Regnault, *Ann. phys chim.*, (Ser. 3), **15**, 1845, (129); *Mém. Acad. Sci.*, **26**, 1862, (679).

[14] Campbell, *Trans. Faraday Soc.*, **10**, 1914, (197).

[15] If these two pressures were very close, and if the gauge tubes were too narrow, the total pressure within the hot gauge might be appreciably greater than the pressure within the reservoir.

[16] Happel, *Ann. Physik*, (Ser. 4) **13**, 1904, (340), etc.

[17] Van Laar, *Verslag. Akad. Wetenschappen*, **24**, 1916, (1635).

[18] Van Laar, *Proc. Acad. Sci. Amsterdam*, **20**, 1917, (138).

[19] Ariès, *Paris, C. R. Acad. Sci.*, **166**, 1918, (334).

[20] "A Lower Limit for the Critical Temperature of Mercury," presented before the Division of Inorganic and Physical Chemistry, 1910.

[21] Koenigsberger, *Chem. Ztg.*, **135**, 1912, (1321).

[22] Menzies, *J. Amer. Chem. Soc.*, **35**, 1913, (1065).

[23] Bender, *Physik. Zr.*, **16**, 1915, (246); **19**, 1918, (410).

COHESION, INTERNAL PRESSURE, ADHESION, TENSILE STRENGTH, TENSILE ENERGY, NEGATIVE SURFACE ENERGY, AND MOLECULAR ATTRACTION

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Communicated by W. A. Noyes, October 14, 1919

Work of adhesion.—For the past seven years I have been engaged in the study of molecular attraction from a new point of view, as represented by a thermodynamic equation which does not seem to have been developed before, in spite of the fact that it is exceedingly simple. It gives what may be called the *total adhesional energy* (E_A), or the total energy involved in the approach of two unlike surfaces. The equation developed by Dupré[1] in 1869 gives the *adhesional work* (W_A), which is

the amount of work involved in the process, which is equal to the decrease of free energy during the approach:

$$W_A = -\Delta\gamma = \gamma_1 + \gamma_2 - \gamma_{1,2} \quad (1)$$

where γ_1 and γ_2 give the free energy of the two unlike surfaces before their approach, and $\gamma_{1,2}$ is the free energy of the interface.

It has been shown by Hardy,[2] and by Harkins, Brown, and Davies,[3] that the adhesional work between organic liquids and water is represented by values which are characteristic for each class of organic compounds. Our own work indicated also that at the interface between water and an organic liquid, any groups in the latter which contain oxygen, nitrogen, or either triple or double bonds, are oriented toward the aqueous phase, while the hydrocarbon radical turns toward the organic liquid. This orientation is of great importance in connection with either the adhesional work or the adhesional energy, since its result is to increase the adhesion between such an organic liquid and water. Further facts which are in accord with this theory of orientation, also developed by Langmuir,[4] are presented later in this paper, in the section on negative surface energy.

Work of surface cohesion.—Since γ is zero when the two liquids become identical, the Dupré equation reduces to

$$W_{SC} = -\Delta\gamma = 2\gamma \quad (2)$$

where W_{SC} represents the work of the surface cohesion. The free surface energy (γ) is usually measured in ergs per square centimeter. It is evident that W_{SC} is the work necessary to break a bar of liquid or solid with a cross section of 1 sq. cm., in such a way as to give two plane surfaces of 1 sq. cm. each, that is the break must occur perpendicular to the long axis of the bar. This amount of work is clearly that which would be done in the determination of the tensile strength of a steel or other metal bar, if the test could be carried out under *ideal* conditions, which are that the bar during the test must remain of uniform cross section, and the break must occur in such a way as to give two surfaces which are as closely plane as the surface of a liquid, though in the actual tests this condition is not met, since the bar is much distorted. The work of surface cohesion may therefore be called the *tensile work* (W_T). This is equal to the *tensile force* integrated through the distance necessary to pull the two surfaces completely apart, or

$$W_T = \int_{S_0}^{\infty} F_T ds = 2\gamma$$

Energy of adhesion.—The equation developed by me gives the *total adhesional energy* (E_A), and is as follows:

$$E_A = -\Delta E_S = (\gamma_1 + l_1) + (\gamma_2 + l_2) - (\gamma_{1,2} + l_{1,2}) \quad (4)$$

The energy of adhesion is equal to the *total surface attraction*, (π'') due to molecular attraction, integrated through the distance necessary to pull the two surfaces completely apart, or

$$E_A = \int_{S_0}^{\infty} \pi'' \, ds = E_1 + E_2 - E_{1,2} \quad (5)$$

The only experimental results which have been obtained in connection with equation 4 are those of experiments by Dr. E. C. H. Davies and Mr. Y. C. Cheng as carried out under my direction. This seems to be the first work aside from that on the heat of adsorption, which bears very directly on the molecular attraction between *unlike* substances.

Energy of surface cohesion.—When the two liquids become identical equation 4 reduces to the form

$$E_{SC} = -\Delta E_S = 2\,(\gamma + l) = 2\,E_S \quad (6)$$

It is evident that the total energy of surface cohesion is equal to the total tensile energy (E_T), or

$$E_T = -\Delta E_S = 2E_S \quad (7)$$

Thus the *total energy* used in pulling a bar of unit cross section apart in such a way as to form two unit plane surfaces, is equal to twice the total surface energy. It is also equal to the force of surface cohesion (F_{SC}) integrated through the distance necessary to pull the two surfaces completely apart, or

$$E_T = \int_{S_0}^{\infty} F_{SC} \, ds$$

Energy of cohesion.—The *cohesional energy* of a solid or a liquid may be defined as the energy which would be liberated in the formation of the solid or liquid from its individual molecules, the molecules in the initial state being placed so far apart that they do not attract each other appreciably. This is equal to the latent heat of vaporization plus the heat absorbed in the expansion of the vapor until it becomes very dilute, minus the external work of vaporization, or it is equal to the internal latent heat of vaporization when the liquid is vaporized at a low pressure.

Internal pressure or cohesion.—The internal pressure of a liquid or a solid has been defined as the rate of transfer of momentum across a unit plane area inside the liquid or solid; and the average force of attraction across this unit area, which is numerically equal to the internal pressure, is the cohesional force, or the *cohesion*.

While the work and total energy of adhesion and of surface cohesion, and the energy of cohesion, may all be obtained from experimental results by the use of simple and exact thermodynamic equations, this is not true of the internal pressure or cohesion. As a matter of fact, there is at present no known means of calculating the cohesion, but there are many methods, which do not agree among themselves, of calculating from inexact equations, values which for various liquids are supposed, when arranged in order of magnitude, to lie in the same order in general as the cohesions themselves. In fact, the cohesion is often defined as equal to a/v^2 the pressure correction term in van der Waals equation. However, since this equation is far from exact in its application to liquids, it is obvious that the cohesion calculated cannot represent at all accurately the internal pressure.

Molecular attraction.—All of the phenomena thus far considered in this paper may be considered as due to the attraction between the molecules in a liquid or a solid. It is customary to consider the molecules as spheres or as points, with the attractive forces dependent upon the distance between the molecules alone, when they are all alike. It has been shown by Harkins, Brown, and Davis,[3] by a measurement of the amounts of energy involved, and by Langmuir[4] by a less direct method, that the forces around different parts of a molecule may be very different in magnitude. Thus in the case of organic compounds the forces are very much higher around any groups containing oxygen, nitrogen, triple, or double bonds, than they are around the hydrocarbon chains. The investigations of Harkins, Grafton and Ewing (these PROCEEDINGS, **5**, 1919, 571) show that if organic substances are arranged according to the magnitude of their adhesional surface work toward mercury, they are not so arranged with respect to water. In this respect the adhesional forces seem to have something of the specific nature which indicates chemical action, and it is well known that the recent work on crystal structure demonstrates that such crystals as those of diamond and of graphite are held together by primary valence bonds. Langmuir[4] considers all cohesional and adhesional forces as chemical, while van Laar[5] has recently published the results of an extensive series of calculations which show that the square root of van der Waals' con-

stant of attraction (a) is additive, and therefore comes to the conclusion that all such forces are physical. The calculations of Einstein,[5] Kleemann,[6] and of Harkins and Clark,[7] have also given coefficients of atomic attraction which are moderately exact constants. Since all of these facts when considered together make it probable that cohesional forces are often less specific than those involved in ordinary chemical reactions, while in many cases they are the same valence forces, it seems to me preferable to use neither of the two words, physical or chemical, and to consider that cohesion is due to electrical and magnetic, or electromagnetic forces, which are probably largely electrical. In a paper [8c] on "An Electromagnetic Hypothesis of the Kinetics of Heterogeneous Equilibrium, the Structure of Liquids, and Cohesion" I have already traced a connection between cohesion and the completeness of the outer or valence shell, of electrons in the atom or the molecule. The cohesion decreases as the completeness of the outer shell of electrons in the molecule increases.

On the relation between cohesion and cohesional and adhesional work and energy.—A number of attempts have been made to calculate the *cohesion*, which is the cohesional force per unit area of the cohesional pressure, from the *cohesional surface work*, and this attempt has been more or less justified by the fact that the values thus obtained, while not good in numerical agreement with those given by a/v^3, lie on the whole in the same relative order. Such calculations have been made by Mathews,[9] and by Hildebrand,[10] but neither of them have shown to what extent the cohesion and the cohesional surface work are related. The term a/v^2 may be said to represent, more or less imperfectly it is true, the total effect of the molecular attraction in decreasing the external pressure, which decrease is the cohesion. The cohesional surface work, on the other hand does not represent the total effect of the molecular attraction, even as it acts in a surface, since the molecules move into the surface not only by means of the energy which is contributed in the form of work, but also by means of the utilization of the kinetic energy of molecular motion, or the latent heat of the surface. Thus the formation of the surface of a pure liquid, with the exception of a few liquids in which liquid crystal formation is involved, is always accompanied by cooling. It is, therefore, the *total cohesional surface energy* and *not the related work*, which represents the total effect of the molecular attraction, or the cohesional effect.

It may seem remarkable, from the point of view of the last paragraph, that the calculation of even the relative cohesion of liquids[10] from a

very simple equation, $\gamma/v^{1/3}$, where V is the molecular volume, should give results which lie in somewhat the correct order. This is undoubtedly because, as shown by Harkins, the contribution of the kinetic energy of a molecule to the total energy of the surface, is on the whole, independent of the nature of the molecule,—at least for such substances as have been used in the calculation of cohesion,—and is dependent on the temperature alone. Therefore, so long as the molecular volume is nearly the same, and the orientation of the molecules in the surface is not an important factor, at any definite temperature the latent heat of the surface is nearly independent of the nature of the substance, so that when substances are arranged in the order of their cohesional surface work or their free surface energy, they are also arranged in the general order of their total surface energy.

A second method of calculating the cohesion from the cohesional work or free surface energy, is based on the well-known assumption of Stefan,[11] that the average work involved in bringing a molecule to the surface of a liquid, has one-half of the energy value involved on the average in its complete vaporization. It is obvious from Stefan's paper, that his principle does not involve the *work* but the *total surface energy*, which is supposed to be one-half of the latent heat of vaporization. That this rule is far from true is indicated by the results of extensive calculations by Mr. L. E. Roberts and myself, which show that the fractional contribution of the surface energy toward the complete vaporization increases with the temperature, with a normal range of from one-third at lower temperatures to 0.8 or more as the critical temperature is approached, though the higher values are uncertain. Thus a molecule which at a high corresponding temperature passes from the body of the liquid into the surface, goes, in a *fractional sense with reference to energy*, much more nearly into the vapor state than when the corresponding temperature is low.

Negative surface energy.—The phenomenon of *negative surface energy* was first discovered two years ago by Dr. E. C. H. Davies and me, but has not been announced previous to this time in print. Not only Donnan, but also Tolman and Wolfgang Ostwald, have *assumed* the existence of a negative surface tension or free surface energy. My own investigations have convinced me that the discovery of a negative free surface energy for a plane, uncharged surface is improbable, though it is quite likely that there is such a phenomenon in the case of highly curved phase boundaries. What we have to announce here is the discovery of *a negative total surface energy for a plane, uncharged sur-*

face. Thus, contrary to the rule found in the past, the surface or interface between octyl alcohol and water *gives off energy when it is extended*, but, nevertheless, the *surface cannot be formed without the expenditure of work*. The apparent contradiction is due to the fact that while the molecular motion aids in the formation of an ordinary surface, in the case of the interface under discussion the molecular motion hinders the extension of the surface. This is in accord with the theory presented in our earlier papers, and by Langmuir, that at such an interface there is an orientation of the molecules, since the molecular motion reduces the extent of the orientation.

When the interface between octyl alcohol and water is pulled out adiabatically there is thus a heating of the surface, while an ordinary surface is cooled, so that the potential energy of the molecules is decreased by passing into the alcohol-water interface. The negative surface energy, is, it is true, very small, with a numerical value of two ergs. per sq. cm., while the free surface energy is 8.33, and the latent heat is -10.3 ergs. In contrast with this, it is found that the total surface energy of the hexane-water interface is not only positive but large, with a value of 66.5 ergs. These relations are of considerable interest, and their bearing on interfacial structure, which is of great importance in physiology, will be discussed in a later paper in the *Journal of the American Chemical Society*.

[1] Dupré, *Theorie Mécanique de la Chaleur*, Paris, 1869, p. 69; Lord Rayleigh, *London, Phil. Mag.*, (5) 30, 1890, (461).

[2] Hardy, *London, Proc. Roy. Soc.*, 86B, 1911, (634).

[3] (a). Harkins, Brown, and Davies, *J. Amer. Chem. Soc.*, 39, 1917, (354–64).

(b). Harkins, Davies, and Clark, *ibid.*, 541–96.

(c). Harkins and King, *ibid.*, 41, 970–92, (1919), these PROCEEDINGS, 5, 1919, (152–9).

[4] Langmuir, *J. Amer. Chem. Soc.*, 39, 1917, (1848–1906), these PROCEEDINGS, 3, 1917, (251–7); abstract in Met. Chem. Eng., 15, 1916, (468). Fraenkel, *Phil. Mag.*, 33, 1917, (297–322).

[5] Einstein, *Leipsig, Ann. Physik.*, 4, 1901, (513).

[6] Kleemann, *Phil. Mag.*, 18, 1909, (39, 491, 901).

[7] Harkins and Clark, unpublished calculations.

[8] van Laar, *Zs. anorg. Chemie*, 104, 1916, (57–156).

[9] Mathews, *J. Physic. Chem.*, 17, (603–28).

[10] Hildebrand, *J. Amer. Chem. Soc.*, 41, 1919, (1067–80).

[11] *Leipsig, Ann. Physik*, 29, 1886, (655).

THE ADHESION BETWEEN MERCURY, WATER, AND ORGANIC SUBSTANCES, AND THE FUNDAMENTAL PRINCIPLES OF FLOTATION

By William D. Harkins

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Communicated by W. A. Noyes, October 14, 1919

The primary purpose of this investigation was to determine the effects of the molecular attraction at the surface of a metal, and to compare these effects with those of the surface of an oxygen compound such as water. Since the flotation process depends upon the preferential wetting and adhesion of gas films on metals, including the heavy sulphides, etc., on the one hand and silica and similar substances on the other, the general principles learned in connection with such a study, should be fundamental for the study of the process. I have been informed by Dr. E. C. Bingham that the adhesion between organic substances and metals is also fundamental with respect to the characteristics of lubricants.

The equation of Dupré[1] and that of Harkins, both of which are based on pure thermodynamics, give us the most accurate means for the study of the effects of molecular attraction at surfaces. The equation of Dupré gives the *adhesional work* done during the approach of 1 sq. cm. of one surface to meet the same area of the other. This is numerically equal to the work necessary to pull the two surfaces apart. The work of approach is also equal to the decrease of free surface energy ($-\Delta\gamma$) during the process which is given by the equation

$$W_A = -\Delta\gamma = \gamma_1 + \gamma_2 - \gamma_{1,2}$$

where γ_1 and γ_2 give the free energy of the two unlike surfaces before their approach, and $\gamma_{1,2}$ is the free energy of the interface.

The equation of Harkins gives the *total energy of approach* (E_A) and this may be called the *total adhesional energy*, as follows:

$$E_A = -\Delta E_S = (\gamma_1 + l_1) + (\gamma_2 + l_2) - (\gamma_{1,2} + l_{1,2})$$

where l represents the latent heat of the surface or interface in ergs per square centimeter. The total adhesional energy is closely related to the molecular surface attraction, while the adhesional work is the tensile force necessary to pull the two surfaces apart, integrated through the distance which they move during separation, but given a negative sign.

The data obtained in this laboratory indicate that the adhesional work between a mercury surface and the surface of an organic liquid is always greater, at least for all substances investigated, than that between the same organic liquid and water, and also greater than that between the organic liquid and itself. This last might be more properly described by the term *cohesional surface work*. A second point of interest is that for about half the organic substances investigated, the *difference* between the adhesional work against mercury and that against water, is nearly constant, with a value between 80 and 92 ergs. This is true for such liquids as the paraffin hydrocarbons, benzene, toluene, the xylenes, carbon tetrachloride, chloroform, and nitrobenzene, so *the work is by no means entirely specific*. On the other hand the adhesional work toward water is specifically high in the case of the alcohols, water and ether, while that toward mercury is very high in the case of the compounds of sulphur, iodine, and bromine, and also oleic acid, which indicates that the specific effects are very marked.

At the interface between organic compounds and water, groups which contain oxygen or nitrogen, double or triple bonds, orient toward the water; at a mercury surface the sulphur, bromine, or iodine, should turn toward the mercury.

In order to illustrate these relations a few of the data obtained in the laboratory are presented in table 1. The experimental work on mercury was done under my direction by Dr. E. H. Grafton and Warren W. Ewing, while that on water was selected from an extensive set of data obtained by Drs. F. E. Brown, G. L. Clark, E. C. H. Davies, Mr. L. E. Roberts, and Mr. Y. C. Cheng.

The well known rule of Antonow[2] states that the interfacial tension between two liquids is equal to the surface tension of the liquid which has the higher surface tension minus that which has the lower surface tension. Where this rule does not hold for an interface organic liquid-water, the discrepancy is explained by Antonow as due to the fact that the interface is between two solutions and not two pure liquids, so the surface tensions used should be those of the solutions or phases, instead of the pure liquids. The Antonow rule sprang from the idea that an interface at the two individual surfaces still exists, but that one acts in a negative sense on the other. This rule is in direct discordance with the theory developed independently by Langmuir and by Harkins, that the molecules in surfaces and interfaces are oriented. This theory need not be discussed here, since the above table clearly proves the Antonow rule to be incorrect for immiscible liquids. Thus his rule if it were applica-

TABLE 1

THE ADHESIONAL WORK BETWEEN MERCURY, WATER, AND ORGANIC LIQUIDS, WITH THE COHESIONAL SURFACE WORK IN ORGANIC SUBSTANCES FOR COMPARISON. (LIQUIDS ARRANGED IN ORDER OF ADHESIONAL WORK TOWARD MERCURY)
(IN ERGS PER SQUARE CENTIMETER)

(1) LIQUID	(2) INTERFACIAL TENSION AGAINST MERCURY	(3) ADHESIONAL WORK AGAINST MERCURY	(4) COHESIONAL WORK AGAINST ITSELF	(5) DIFFERENCE (3) MINUS (4)	(6) ADHESIONAL WORK AGAINST WATER	(7) DIFFERENCE MERCURY MINUS WATER (3–6)
(Air)	465					
Hexane	378	120	36.9	83	40	80
Ethyl ether	379	123	43.6	79	73	50
Octane	375	127	43.5	83	44	83
Carbon tetrachloride	362	150	53.3	97	56	94
Chloroform	357	155	54.3	101	67	88
Benzene	363	146	57.6	88	67	79
Toluene	359	151	58.0	93	67	84
m-Xylene	357	152	58.0	94	64	88
o-Xylene	359	153	58.0	95	67	86
p-Xylene	361	155	54.0	101	64	91
Iso-butyl alcohol	348	155	45.6	109	94	61
Secondary octyl alcohol	348	159				
Octyl alcohol	352	161	55.1	106	92	69
Methylene chloride	341	169	53.0	116	71	98
Ethylidene chloride	337	174	49.2	125		
Nitrobenzene	350	173	86.8	86	91	82
Carbon bisulfide	336	175	62.8	113	56	119
Aniline	341	181	85.2	96	110	71
Water	375	182	145.6	36	145.6	36
Oleic acid	322	191	65.0	136	89.6	101
Ethyl iodide	322	195	56.4	139	63	132
Ethylene bromide	326	197	77.4	120	75	122
Methyl iodide	304	211				
Acetylene tetrabromide	293	230	99.3	131	84	146
Mercury		960	960.0		182.6	(778)

ble to the pure liquids would state that the interfacial tension between mercury and acetylene tetrabromide is 430, while the experimental value is 293, an extremely great deviation. Taking another example at random it is found that the Antonow rule in this form gives the mercury-octyl alcohol interfacial tension as 452, while the experi-

mental value is 352. As a matter of fact, the deviation between the result given by rule and by experiment seldom falls below 90 ergs, and is often higher, which seems to disprove the rule.

In spite of the above objections it must be admitted that Antonow's rule has been of considerable use in connection with phase boundaries between water and organic liquids. It should be realized, too, that the above objections to it involve a certain hypothesis, that is that the amount of the organic liquid dissolved in the mercury is too small to reduce the surface tension by 90 ergs or more when the mercury phase is split apart in such a way as to form surfaces from the interior part of the mercury phase.

Preliminary experiments on the adsorption of benzene and other vapors on a mercury surface show that their positive adsorption is very much greater than on a water surface at the same concentration of vapor. Water present either in the air or in such organic liquids as hexane, benzene, ether, etc., is also adsorbed positively, and the adsorption is very marked even when only a very small amount of water is present. Mercury salts, such as mecurous iodide or bromide, when dissolved in an organic iodide or bromide, are strongly adsorbed. The high adsorption of such substances on mercury as compared with that on water is in accord with my theory that the adsorption increases in general with the rate of drop in the intensity of the electromagnetic intermolecular stray field at the surface of the liquid, since this drop is very much more rapid for mercury than for water. According to the results obtained by F. Schmidt[3] caesium, rubidium, and potassium, when dissolved in mercury are strongly, and sodium, tin, lead, and gold, are slightly positively adsorbed. Negative adsorption is most marked with barium as a solute, and to a lesser extent with strontium, calcium, cadmium, zinc, thallium, and lithium, in decreasing order of effect.

The experimental difficulties in connection with the investigation of the surface of a liquid metal are considerable, and they have been solved thus far by Dr. Grafton and Mr. W. W. Ewing. The result which is used in table 1 for the surface tension of mercury in a vacuum, is not our own, and may be too high, but this would not affect the value of the results, since it introduces only a small constant difference in all of the numbers which represent the work of adsorption. The liquids were purified with great care. Many supposedly pure organic substances which are not sulphur compounds, and which have the correct boiling points, contain enough organic sulphides to make determinations made upon them entirely useless. This was especially true of the xylenes, which were boiled with mercury for several days to remove the sulphur.

Experiments have been made on the adsorption of organic acids from aqueous solution, and the results finally obtained, together with those on soaps, all of which have a high positive adsorption, will be reported in the complete papers which will be published in the *Journal of the American Chemical Society*. A recent paper by Hardy[4] gives valuable information on the application of the principles of surface action to the problem of lubrication. Hardy was the first worker to investigate the application of the Dupré equation to interfaces between water and organic liquids. Our complete papers will consider the relation between adhesion and lubrication. Experiments are now in progress on the heat of adsorption of liquids on solids, both on dispersiod and plane surfaces, and calculations are in progress which will relate the results given in this paper to the atomic and molecular distances.

[1] Dupre, *Theorie Mécanique de la Chaleur*, Paris, 1869, p. 69; Lord Rayleigh, Phil. Mag. (5) **30**, (1890), (461), Hardy, *Proc. Roy. Soc., London*, (A) **88**, 1913, (303–33), Harkins, Brown, and Davies, *J. Amer. Chem. Soc.*, **39**, 1917, (354–64).

[3] Antonow, *J. Chem. Phys.*, **5**, 1907, (384).

[3] F. Schmidt, *Leipzig, Ann. Physik*, **39**, 1912, (1108).

[4] Hardy, *Phil. Mag.*, May, 1919.

A BENIGN TUMOR THAT IS HEREDITARY IN DROSOPHILA

By Mary B. Stark

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Communicated by T. H. Morgan, October 16, 1919

1. A non-lethal hereditary tumor.—In a strain of flies with a lethal tumor, i.e. a tumor occurring in one-half of the males and causing their death (*J. Canc. Res.*, July, 1918; *J. Exper. Zool.*, February, 1919) another tumor has appeared as a mutation. The new tumor differs from the lethal one in that it is not sex-linked, i.e., it appears in females as well as males, and further in that it does not cause the death of the flies in which it occurs.

After several generations of inbreeding of males and females with tumors, a stock was obtained which breeds true to the tumor—the tumor appearing in all the flies.

Since the new tumor is not sex-linked its gene is not located in the X chromosome. To locate the gene in one of the other chromosomes, females with tumors were mated to star dichaete males. The gene for

star is located in the second chromosome and that for dichaete in the third chromosome. In the F_2 generation tumors reappeared as frequently among the star as among non-star flies. On the other hand none of the dichaete flies had tumors. This result indicates that one at least of the genes essential for tumor development is in the third chromosome closely linked to dichaete. To locate the gene at a definite locus in the third chromosome, flies with tumors were mated to dichaete 'hairless.' In the back crosses, no crossing over between the tumor and dichaete was obtained, while the crossing over between the tumor and hairless was the normal amount, 25%. The gene of the tumor is thus shown to be very close to that of dichaete which is at about 11.7 (table 1). The expected number of tumor bearing flies in the back cross is 50%. As seen in table 1 not more than 5% developed the tumor. This indicates

TABLE 1

DATE	WILD TYPE		HAIRLESS DICHAETE		HAIRLESS		DICHAETE		HAIRLESS		TUMOR	
	♀	♂	♀	♂	♀	♂	♀	♂	♀	♂	♀	♂
July 10, 1918	13	13	26	24	7	5	10	5	0	2	1	0
	28	25	58	54	9	7	14	9	2	3	3	0
	37	27	28	20	4	3	5	5	4	3	1	1
	17	16	19	17	3	4	9	5	2	2	0	1
	16	14	26	20	4	2	4	4	2	1	1	0
	32	38	30	26	14	16	20	18	4	5	2	0
Total	143	133	187	161	41	37	62	46	14	16	8	2

Crossing over between tumor and hairless = 10/40 = 25%.

that more than one gene is concerned with the inheritance of the tumor. Further investigations of this point are being made.

2. Position of tumor in larva.—To determine the time of appearance of the tumor and its position in the body 208 larvae were isolated and examined for tumors. Tumors were found in 180 of these (table 2). They seem to occur more often in the twelfth and thirteenth segments, occurring in the twelfth 97 times and in the thirteenth 72 times. Two tumors were observed in each of thirty-eight larvae. Twenty-seven of these cases had the tumors in different segments and eleven in the same segment. One larva had three tumors. Twenty-five larvae with tumors had in addition smaller tumors which were regarded as metastases. These when very small are often carried into the heart with the blood and there develop into narrow elongated tumors as shown in figure 1.

The tumor may develop in the early or in the late larval stages. Fully developed tumors have been found in two-day old larvae and very young tumors in four-day old larvae. Of the 208 larvae isolated, only 32 were without tumors. Tumors must have developed later in these individuals since all of the flies that emerged from them had tumors.

One hundred and seventy-six flies emerged from the 208 larvae isolated, approximately the same percentage as in the control experiment in which 200 larvae from a normal stock were isolated, of which 170 emerged as flies.

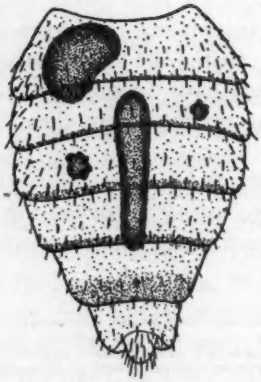
TABLE 2

POSITION OF TUMORS IN SEGMENTS	NUMBER OF LARVAE WITH TUMORS	NUMBER WITH ONE TUMOR	NUMBER WITH TWO TUMORS	NUMBER WITH THREE TUMORS	NUMBER WITH METASTASES
14	5	5			
14–13–12	1			1	
13	49	44	5		6
13–12	16		16		4
13–11	2		2		
13–10	1		1		
13–8	3		3		
12	75	65	10		12
12–11	4		4		1
12–8	1		1		1
11	8	6	2		
9	9	9			
8	5	5			
6	1	1			1
Total.....	180	135	88	3	25

Total number of larvae, 180. Total number of tumors, 226. Total number of metastases, 36.

3. Development of tumor.—The structure of the tumor in an early stage in its development in the larva is shown in figure 2. The cells are rounded or polygonal in shape and contain pigment. As the tumor grows older the amount of pigment increases, the cells filled with it become crowded towards the periphery and flattened as shown in figure 3.

The tumor takes its origin in groups of cells similar in structure to the cells found just inside of the hypodermis of the larva. These cells are originally derived from the hypodermal cells. In the fly the tumor when fully matured is entirely permeated by pigment and is black in color. The cells have stopped increasing in number. As the fly grows older the tumor tends to shrink in size but does not disappear.



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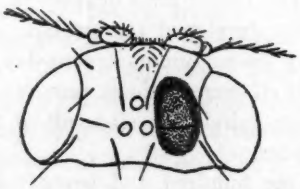

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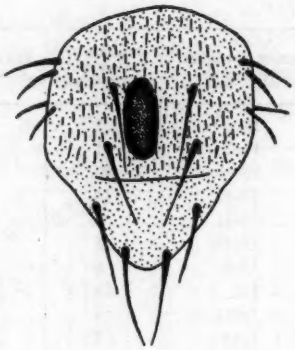

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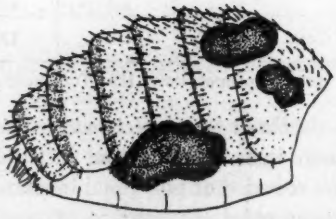

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All figures were drawn with the aid of a camera lucida, using a 16 mm. objective and ocular no. 5 with tube length of 165 mm. excepting figures 2, 3, and 5 for which the 4 mm. objective was used.

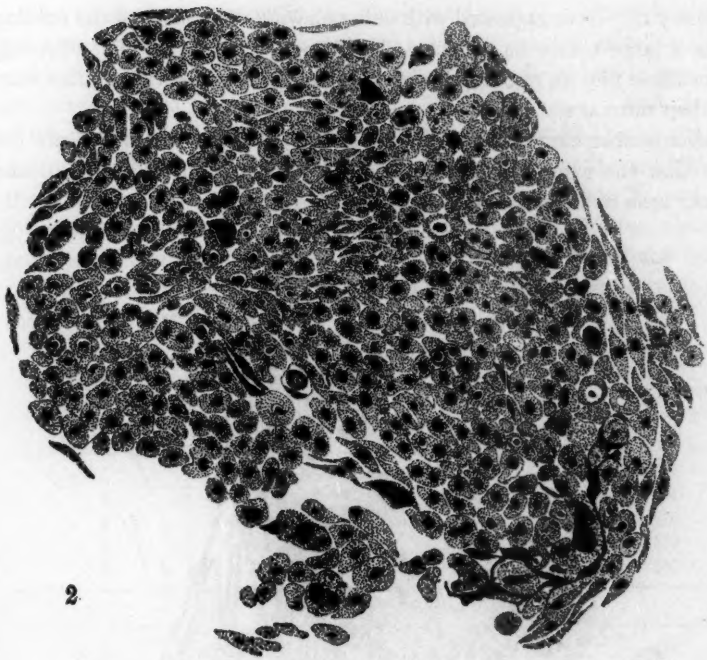
FIG. 1. Dorsal view of abdomen showing a tumor in dorsal artery.

FIG. 4. Dorsal view of head, thorax and first segment of abdomen showing a tumor in the place of the left wing.

FIG. 6. Dorsal view of head with a tumor.

FIG. 7. Dorsal view of thorax with one tumor.

FIG. 8. Side view of abdomen with three tumors.



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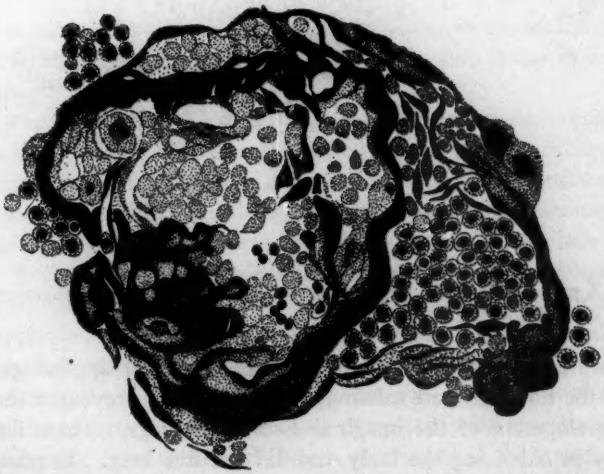

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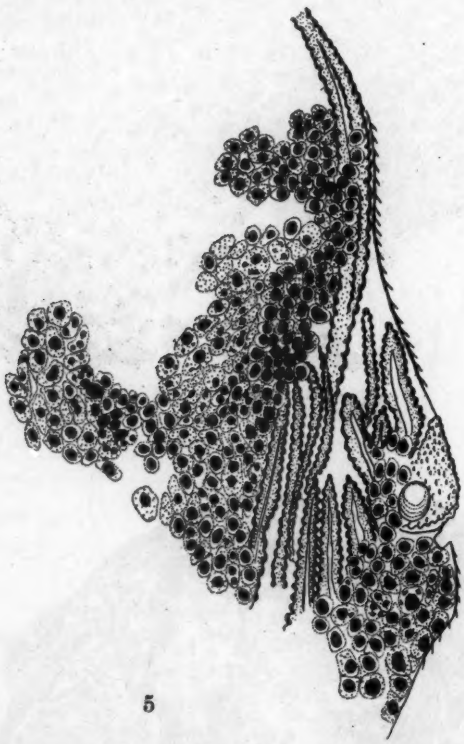
FIG. 2. A section through the center of a tumor in an early stage of development.
FIG 3. A section through the center of a tumor in a late stage of development.

Many flies have appeared with only one wing. In place of the missing wing a large tumor has developed as shown in figure 4. The offspring from these flies do not throw a greater number of one-winged flies than do their normal sisters.

Microscopic examination of sections of the wing tumor reveals the fact that the wing disc had begun to develop but an ingrowth of the tumor cells had checked the development (figure 5).



5

FIG. 5. A section through the wing tumor (fig. 2) showing ingrowth of tumor cells into the partly developed wing disc.

Tumors have also appeared in place of the other appendages. In several, the fore-legs were missing, a tumor having prevented the complete development of the imaginal discs of the legs. These flies had difficulty in balancing the body and did not live long. In many flies only one leg or only a portion of the leg would be displaced by a tumor.

The tumor may appear in the head, see figure 6. Many flies with head-tumors seemed to have no power of coordination and lived only a few days, while other flies did not seem to be disturbed in any way whatever.

When the tumor developed in the abdomen or in the thorax (figs. 1, 7 and 8) and did not interfere with the development of the appendages the length of life of the fly seemed to be normal. Table 3, part 'a,' gives the length of life of the females of five pairs of flies, also the number of offspring from each pair. The length of life of the female and the number of offspring correspond closely with same of control as indicated in part 'b' of table 3.

TABLE 3

NUMBER OF PAIRS	LENGTH OF LIFE OF ♀	NUMBER OF DAUGHTERS	NUMBER OF SONS
a. Pairs of flies having tumors (February 30, 1919)			
1	18 days	296	257
2	18 days	264	271
3	19 days	198	110
4	36 days	154	139
5	16 days	154	163
b. Control pairs of normal flies			
1	20	304	264
2	35	263	232
3	22	198	207

It is evident that the presence of the tumor in the abdomen and in the thorax of the body is not sufficiently injurious to decrease seriously the length of life of the fly.

4. Inoculation of tumors.—Under aseptic conditions young tumors were removed from larvae and inserted into larvae of a different strain normally free from tumors. Forty larvae were thus inoculated. Only five percent (two) survived the operation, but these continued to live, completed metamorphosis and carried the inserted tumor into the adult fly. Both these flies were females and were sterile. Matings were attempted with several males but without success. The tumor was found to resemble exactly the tumor that occurs normally in the adult fly in the stock from which it was originally removed.

A few of the larvae from which the tumors were removed recovered from the operation. Some of these completed metamorphosis. All of the offspring from these had tumors showing that the tumor is due to something in the germ-plasm which is handed over from one generation

to another even though the parents are deprived of the tumors by an operation.

I am gratefully indebted to Professor Morgan and Doctor Bridges for helpful suggestions.

Conclusions.—1. A non-lethal tumor appeared as a mutation in the lethal tumor strain.

2. The locus of the gene of the new tumor is close to that of the dichaete in the third chromosome.

3. The tumor may occur in any segment of the larva but seems to occur more often in the twelfth and thirteenth segments.

4. The cells of the tumor are rounded or polygonal in shape and show the presence of pigment.

5. Ingrowth of tumor cells into the imaginal discs of the appendages checks the development of the parts.

6. Young tumors were inserted into larvae of normal strains. Five per cent survived the operation, completed metamorphosis and carried the inserted tumor into the adult fly.

METALLIC SALTS OF PYRROL, INDOL AND CARBAZOL

By Edward C. Franklin

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Communicated, October 20, 1919

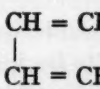
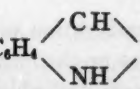
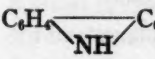
The ammono acids, that is to say, the acids of the ammonia system of acids, bases and salts,[1] are derivatives of ammonia in which one or two hydrogen atoms of the ammonia molecule are replaced by negative groups. A number of examples of compounds so related to ammonia are nitramide or nitrosyl amide, NO_2NH_2; acetamide or acetyl amide, CH_3CONH_2; phthalimide or phthalylimide, $C_6H_4\langle{}^{CO}_{CO}\rangle NH$; benzenesulfonnitramide or nitrosyl benzenesulfonyl imide, $C_6H_5SO_2NHNO_2$; methylnitramine or methyl nitrosyl imide; acetanilide or phenyl acetyl imide; trinitraniline, $C_6H_2(NO_2)_3NH_2$; cyanamide, $CNNH_2$; urea, $CO(NH_2)_2$ etc., etc.

These substances are true acids ranging in acidity from benzenesulfonnitramide, which approaches the ordinary mineral acids in strength, through phthalimide and methyl nitramine, which are well known to possess weak acid properties, to acetamide and urea which are not

ordinarily recognized as acids at all. The acid properties of acetamide, urea and other very weak ammono acids, however, show themselves distinctly when in solution in liquid ammonia as has been shown by the writer and his students in earlier papers.

The fact that acids too weak to be recognized as such in aqueous solution are still capable of showing acid properties when in solution in liquid ammonia is undoubtedly due to the lower solvolytic[2] action of ammonia as compared with water which in turn results from the very slight autoionization of the former solvent. To what extent liquid ammonia is dissociated has not been accurately determined. Since, however, it is a comparatively easy matter to obtain liquid ammonia with a specific conductance less than one-eighth that of water[3] it must be considerably less ionized than is the latter solvent.

It happens therefore that salts of the weaker ammono acids, which, because of the strong hydrolytic action of water are incapable of existence in the presence of this solvent, have been easily prepared from liquid ammonia solutions.

Pyrrol,  , indol,  and carbazol,  , are obviously derivatives of ammonia in which ammonia hydrogen is replaced by distinctly negative groups. They are therefore to be classed among the ammono acids and as such should react, in liquid ammonia solution, with the more electro positive metals and with their amides, the ammono bases, to form salts.

Experiments will be described in a paper to be printed in the *Journal of Physical Chemistry* showing that pyrrol, indol and carbazol react in liquid ammonia solution with metallic potassium, sodium, calcium and magnesium and with the amides of potassium, sodium, calcium and silver to form the following well defined salts:

Sodium pyrrol, C_4H_4NNa and $C_4H_4NNa.NH_3$.
Calcium pyrrol, $(C_4H_4N)_2Ca$ and $(C_4H_4N)_2Ca.4NH_3$.
Magnesium pyrrol, $(C_4H_4N)_2Mg.2NH_3$.
Silver pyrrol, $C_4H_4NAg.NH_3$.
Sodium indol, $C_8H_6NNa.xNH_3$.
Potassium indol, $C_8H_6NK.xNH_3$.
Calcium indol, $(C_8H_6N)_2Ca.4NH_3$.
Magnesium indol, $(C_8H_6N)_2Mg.4NH_3$.

Silver indol, $C_8H_6NAg.NH_3$.
Potassium carbazol, $C_{12}H_8NK.2NH_3$ and $C_{12}H_8NK.NH_3$.
Calcium carbazol, $(C_{12}H_8N)_2Ca.7NH_3$ and $(C_{12}H_8N)_2Ca.4NH_3$.
Silver carbazol, $C_{12}H_8NAg.2NH_3$ and $C_{12}H_8NAg.NH_3$.

[1] *Amer. Chem. J.*, **28**, 1902, (83); **47**, 1912, (285); *Eighth Int. Cong. App. Chem.* **6**, 1912, (119) and *J. Amer. Chem. Soc.*, **37**, 1915, (2279).

[2] Solvolysis is used as a general term to include hydrolysis, ammonolysis, amidolysis, aminolysis, alcoholysis, etc.

[3] Franklin and Kraus, *J. Amer. Chem. Soc.*, **27**, 1905, (191).

GROWTH AND REPRODUCTION IN FOWLS IN THE ABSENCE OF CAROTINOIDS AND THE PHYSIOLOGICAL RELATION OF YELLOW PIGMENTATION TO EGG LAYING

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Communicated by W. A. Noyes, November 1, 1919

The chemical identification of each of the recognized vitamines as individual substances or chemical groups is greatly to be desired. As the result of certain studies[1] which I made on the physiological relation between the yellow carotin and xanthophyll pigments of plants and the yellow lipochromes of animal tissues and fluids, I became impressed with the fact that there seemed to be more than a casual relation between the simultaneous presence of the plant carotinoids and fat-soluble vitamine in butter fat and egg yolk and in the leafy parts of green plants, and the simultaneous absence of carotinoids and fat-soluble vitamine from lard.

That phase of my carotinoid studies showing the physiological identity of egg yolk lipochrome with plant xanthophyll suggested that the fowl should be a suitable animal upon which to test the relation of plant carotinoids to growth and reproduction. It was decided to approach the question by attempting to raise a flock of chickens from hatching to maturity on a ration devoid of carotin and xanthophylls. The problem which was presented was therefore mainly one of selecting a ration devoid of yellow plant pigments but which was presumably adequate otherwise for the normal growth of chickens.

Three experiments were undertaken. The first was a preliminary experiment, during the winter of 1916–17, to test the efficacy of a ration of white corn, white corn bran, bleached flour, skim-milk and

bone meal, beginning with birds weighing about one-half pound each. Several birds reached the period of fecundity on this ration devoid of carotinoids, and the yolks of the eggs laid were remarkably deficient in color.

In the second experiment, carried out during the season of 1917–18, 75 White Leghorn chicks were placed on a practically identical carotinoid-free ration immediately after hatching. None of these birds reached maturity, partly, it is believed, because of nutritional difficulties, and partly, it is admitted, because of a number of unfortunate accidents in caring for the birds, which caused the death of the larger share of them.

The nutritional difficulties of the second experiment were overcome in the third trial begun in April, 1918, by introducing into the ration both pork liver, which is rich in vitamines,[3] but devoid of carotinoids,[3] and a roughage of paper pulp.[4] A flock of 50 vigorous, normal White Leghorn chickens were raised from hatching to maturity on this modified white corn and skim-milk ration. The mature birds were free from yellow pigmentation.

Not only was normal growth secured on this ration, but the hens in the flock exhibited normal fecundity. Seventeen of the hens whose egg records were kept averaged 52 eggs each during a period of 233 days. Several had considerably higher records. One hen laid 44 eggs during a period of 59 days.

Especially interesting was the character of the pigmentation of the egg yolks. The hard-boiled yolks were colorless but the raw yolks had a faintly yellow color. The pigment, however, was neither carotin nor xanthophyll. Acetone readily extracted the coloring matter from the raw yolks but the pigmented fat which could be obtained from this extract failed to give the reduction test with ferric chloride, which I have shown[5] to be characteristic of carotinoids. Attempts to identify the pigment with Barbieri's[6] ovochromine or with bilirubin, were not successful.

It was felt that the negative relation between carotinoids and fat-soluble vitamine as exhibited by normal growth and fecundity in chickens on rations devoid of carotinoids could not be regarded as established unless the carotinoid-free eggs should prove fertile and normal chicks be hatched from them. Inasmuch as the cocks and hens of the carotinoid-free flock were kept together throughout the experiment the eggs were presumably fertile. About 90 eggs, in all, were incubated at various times. Forty-one livable chicks were hatched

from these eggs. The young chicks appeared normal in every way except for the complete absence of yellow pigmentation from the shanks, beaks, and other skin parts.

The newly hatched chicks were immediately placed on a carotinoid-free ration and were cared for as nearly as possible in the same manner as the young chicks of the preceding generation. By the end of three months, however, all had died. Although it is probable that this unfortunate result may be explained on the ground that the chicks were hatched very late in the season and therefore had to combat a period of extreme heat, as well as a very restricted diet during the most precarious period of their growth, nevertheless the question remains open as to whether it is possible to continue the carotinoid-free condition into more than one generation.

Physiological relation of pigmentation to egg laying.—Practical poultry men have recognized for several years that a relation exists between the amount of yellow pigment visible in the shanks, ear lobes, beak and vent of hens of the Leghorn, Plymouth Rock, Wyandotte and Rhode Island Red breeds and their previous egg laying activity. Extensive biometric analyses have been made by Blakeslee and Warner[7] and by these authors with Harris and Kirkpatrick[8] of data collected at the Storrs Agricultural Experiment Station egg laying competitions in order to establish the character of this relation. The results show a positive correlation between pale colored shanks, ear lobes, beak, etc., and a recent more or less heavy egg production.

The hypothesis which has been adopted by these investigators to explain the physiological relationship which has been observed between fecundity and pigmentation is that the growth of the egg abstracts the pigment from the body tissues. The idea that the relationship could be explained also on the basis that the egg yolk abstracts fat-soluble pigment from the food, thus precluding its localization in the body tissues, was advanced by Harris, Blakeslee and Warner[9] in an earlier paper, but was apparently abandoned. The high percentage of fat in the blood of laying hens, as compared with non-laying hens, as shown by Warner and Edmonds,[10] and by Riddle and Harris,[11] is believed by the former authors to support the hypothesis that the tissue fat is being transferred to the egg yolk during laying with a consequent subtraction of pigment.

The success attained in raising a flock of White Leghorn fowls entirely lacking in pigmentation in both adipose tissue and visible skin parts presented the opportunity for ascertaining the true physiological relation

between fecundity and the fading of the yellow pigmentation of the shanks, ear lobes, etc. The fact that the carotinoid-free hens exhibited normal fecundity enhanced greatly their value for the investigation.

The question was attacked in two ways, first, by observing the histological changes in the shank skin when carotinoid-free food was fed to non-laying pigmented birds, and second, by observing the effects on the tissue and skin pigmentation of feeding carotinoid-rich food to laying carotinoid-free hens. The birds used for the histological studies comprise several yellow shanked White Leghorn cockerels, the specific source of whose pigmentation was not known, and one cockerel from the carotinoid-free flock whose feed was changed from the carotinoid-free ration to one composed principally of yellow corn. The visible skin parts of the latter bird took on a yellow color very rapidly after the introduction of the yellow corn until at the end of 42 days his plumage had a rich creamy appearance and the shanks, beak, ear lobes and vent a deep yellow color. Each of the pigmented birds was placed on a carotinoid-free ration and histological studies made on vertical frozen sections of the shank skin of individuals from time to time as the pigment gradually faded.

As the result of these studies the observation of Barrows[12] was confirmed that the yellow pigment of the shank skin is confined chiefly to the Malphigian layer of the epidermis, with some pigment in the corium. Especially instructive were the sections after staining with Nile blue. The sample of this dye which was used was found to be dichromatic with respect to fat and pigment, fat staining red and carotinoid pigment deep blue. By this means it was determined that carotinoid pigment exists free in granular condition in the shank epidermis, which is contrary to the results reported by Barrows, who concluded that the lipochrome of the shank skin is dissolved in fat. The failure of Sudan III to color the visible skin parts of fowls, as observed by Blakeslee,[13] and confirmed by me, is explained readily by the observation that the Malphigian layer of both the pigmented and non-pigmented skin lacks appreciable amounts of stainable fat.

The histological studies of the shank skin as the xanthophyll gradually faded on the carotinoid-free ration showed first a disappearance of pigment from the corium, then a disappearance from the outer layer of the corium which gradually extended to the retè of Malphigi, the last pigment to disappear being the xanthophyll at the base of the Malphigian layer. These observations are interpreted to mean that when

the supply of xanthophyll for the skin is cut off by reason of its removal from the food, or for any other reason, any xanthophyll present in the corium layer of the skin of the shank, ear lobes, etc., is deposited in the rete of Malphigi. At the same time the xanthophyll deposits in the outer layer of the epidermis either wear off by reason of the normal replacement of the outer cells by those lower down, or are oxidized because of closer contact with the air. The xanthophyll deposits in the rete of Malphigi in time become a part of the outer layers of epidermis and are lost also. The skin thus finally becomes free from visible yellow pigment.

The significance of this interpretation at once becomes apparent in the light of the results secured when xanthophyll-rich rations were fed to the laying carotinoid-free hens. After a month on rations containing an abundance of green feed or yellow corn not a trace of xanthophyll had appeared in the ear lobes, shank or vent, and the adipose tissue had taken up such a small amount of yellow color that a very careful examination of the rendered, melted, body fat was necessary to detect the increase in color in comparison with the fat from birds which had received no carotinoids in their ration from birth. The blood serum and the yolks of the eggs laid during the feeding of the xanthophyll-rich rations, however, contained an abundance of yellow pigment.

As the result of the histological studies and feeding trials the author believes that the correct explanation of the physiological relation between egg laying and the fading of visible yellow pigmentation from the bodies of fowls of certain breeds is that in cockerels and non-laying females the visible skin parts represent a normal path of excretion of the xanthophyll pigment derived from the food. Egg laying deflects the excretion entirely to the ovaries, and even prevents the incorporation of xanthophyll with the tissue fat, and this continues as long as the ovaries function with regularity, whether the egg production be at the rate of one egg a day or one egg a week. The result is that the pigment found in the skin at the onset of fecundity is gradually excreted toward the epidermis where it either wears away as the result of the normal structural changes in the epidermis, or becomes oxidized, and thereby decolorized. The movement of yellow skin pigment during fecundity is thus outward and not inward toward the ovaries.

Influence of various feeds and certain dyes on the color of the egg yolk and body fat.—A critical study was made of the effect of certain coloring matters on the pigmentation of adipose tissue, egg yolk and visible skin parts, and also of the relative xanthophyll content of various

materials commonly used as chicken feed, using the carotinoid-free flock.

Carotin alone, fed as naturally highly colored butter fat, and the orange-yellow pigment of the annatto seed, were found to be without influence on the color of the adipose tissue or visible skin parts. Sudan III colored only the adipose tissue of non-laying birds, the visible skin parts being unaffected by this dye. With laying birds, the egg yolk as well as the adipose tissue was colored by Sudan III, but not the visible skin parts.

Yellow corn and green feed only were found to be rich in xanthophyll when various plant and animal materials were tested for their xanthophyll content by their effect on the color of the egg yolks when fed to hens laying carotinoid-free eggs. A little of the pigment was found in hempseed, barley, gluten feed and red corn. Wheat, wheat bran, oats, cottonseed meal, rape seed, meat scraps, blood meal, skim-milk and butter-milk were found to contain negligible quantities of xanthophyll.

The experiments here reviewed were conducted at the Missouri Agricultural Experiment Station with the coöperation of Prof. H. L. Kempster of the Department of Poultry Husbandry. The complete data appeared in a recent issue of the *Journal of Biological Chemistry*.

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[2] Osborne, T. B. and Mendel, L. B., *J. Biol. Chem.*, **32**, 1917, (309–323); **34**, 1918, (17–27).

[3] Palmer, L. S., *Ibid.*, **27**, 1916, (27–32).

[4] Osborne, T. B. and Mendel, L. B., *Ibid.*, **33**, 1918, (433–438).

[5] Palmer, L. S. and Thrun, W. E., *J. Ind. Eng. Chem.*, **8**, 1916, (614–618).

[6] Barbieri, N. A., *Paris, C. R. Acad. Sci.*, **154**, 1912, (1726–1730).

[7] Blakeslee, A. F. and Warner, D. E., *Science*, **41**, 1915, (432–434); *Amer. Nat.*, **49**, 1915, (360–368).

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[9] Harris, J. A., .Blakeslee, A. F. and Warner, D. E., These PROCEEDINGS, **3**, 1917, (237).

[10] Warner, D. E. and Edmond, H. D., *J. Biol. Chem.*, **31**, 1917, (281–294).

[11] Riddle, O. and Harris, J. A., *Ibid.*, **34**, 1918, (161–174).

[12] Barrows, H. R., *Maine Agric. Exp. Sta. Bull.*, No. 232, 1914, (237–252).

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RADIATIONLESS ORBITS

By Edwin Bidwell Wilson

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Read before the Academy, November 10, 1919

The reaction on an electric charge due to non-uniform motion has been calculated (by Lorentz, for example) and found to be

$$\mathbf{F} = \frac{e^2}{6\pi c^3}\frac{d^2\mathbf{v}}{dt^2} \tag{1}$$

this term $\mathbf{F}$ is merely the first of a series which converges with extreme rapidity provided the dimensions of the charge are small compared with the distance light would travel in the time required for a sensible charge in the velocity.[1] The activity of the force $\mathbf{F}$ determines the rate R of radiation of mechanical energy as

$$-R = \mathbf{F}\cdot\mathbf{v} = \frac{e^2}{6\pi c^3}\mathbf{v}\cdot\frac{d^2\mathbf{v}}{dt^2} \tag{2}$$

What plane orbits are such as to make the radiation as determined by (2) identically zero? The answer to this question is contained in the general integral of the quadratic differential equation of the third order

$$\mathbf{v}\cdot\frac{d^2\mathbf{v}}{dt^2} = \frac{dx}{dt}\frac{d^3x}{dt^3} + \frac{dy}{dt}\frac{d^3y}{dt^3} = 0. \tag{3}$$

The integration may be obtained easily by considering $\mathbf{v}$ as the radius vector in the hydrograph. Then $d^2\mathbf{v}/dt^2$ is the acceleration of the moving point in the hydrograph and its radial and tangential components are

$$\frac{d^2v}{dt^2} - v\left(\frac{d\varphi}{dt}\right)^2 \quad \text{and} \quad \frac{1}{v}\frac{d}{dt}\left(v^2\frac{d\varphi}{dt}\right)$$

of φ be the inclination of $\mathbf{v}$ to a fixed direction. The condition of perpendicularity (3) may thus be written

$$\frac{d^2v}{dt^2} - v\left(\frac{d\varphi}{dt}\right)^2 = 0, \qquad \text{or} \qquad \frac{d\varphi}{dt} = \sqrt{\frac{v''}{v}}.$$

Then

$$x = \int [v\cos \int \sqrt{v''/v}\, dt]\, dt, \tag{4}$$

$$y = \int [v\sin \int \sqrt{v''/v}\, dt]\, dt$$

are the parametric equations of the path, v being any function of the time. The equations contain three constants of integration, which allow an arbitrary choice of axes, and one arbitrary function v. The accelerations along and perpendicular to the path are v' and $\sqrt{vv''}$; the total acceleration is $\sqrt{v'^2 + vv''}$, and its inclination to the path, $\tan^{-1}(\sqrt{vv''}/v')$.

A simple case may be had by taking $v = e^{-at}$, which represents a particle combing to rest. Equations (4) may be written

$$x = \int_{\infty}^{t} e^{-at} \cos at\, dt, \qquad y = \int_{\infty}^{t} e^{-at} \sin at\, dt.$$

Or

$$x = \frac{1}{\sqrt{2}\, a} e^{-at} \cos (at - \tfrac{3}{4}\pi), \qquad y = \frac{1}{\sqrt{2}\, a} e^{-at} \sin (at - \tfrac{3}{4}\pi).$$

And if

$$\theta = at - 3\pi/4, \qquad r = \frac{1}{\sqrt{2}\, a} e^{-\theta - 3\pi/4}$$

is the polar equation of the locus—showing an equiangular spiral with 45° between radius and tangent. If $z = x + iy$,

$$z = \frac{1}{\sqrt{2}\, a} e^{-a(1-i)t - 3\pi i/4} \tag{5}$$

The vector velocity dz/dt is 135° ahead of the radius z, and the acceleration is 270° ahead, which means a retarding acceleration decreasing the areal velocity and perpendicular to the radius. This type of acceleration is found in the straight-bore centrifugal gun.

In (5) the magnitudes of the radius $\mathbf{r}$, velocity $\mathbf{v}$, acceleration $\mathbf{v}'$, and its rate $\mathbf{v}''$ are in geometric progression with the ratios $\sqrt{2}\, a$, the value of $\mathbf{v}'$ being $\sqrt{2}\, ae^{-at}$. If this acceleration be resolved along the radius and along the normal to the path, the respective components are $2\sqrt{2}ae^{-at}$ outward and $2\, ae^{-at}$ inward. A constant magnetic field perpendicular to the plane of motion is competent to furnish a component acceleration along the normal to the path and proportional to the velocity. The electric field in a plane perpendicular to the line joining two like charges at its middle point will act radially from the point of equilibrium upon a like charge with a force proportional to the distance. A proper slight adjustment of the intensity of the magnetic field will take care of the force (1). It is therefore possible easily to set up an electrical problem which is satisfied by the path (5). The general dynamical equation for the motion thus adjusted is

$$\frac{d^3z}{dt^3} - 2a\,(1 + aA)\,i\,\frac{dz}{dt} - 2a^2z = A\,\frac{d^3z}{dt^3}, \tag{6}$$

and has the roots $a(1 - i)$ and $1/A$ in addition to $-a(1 - i)$; two of the roots therefore give increasing motions. The root $1/A$ is the large extraneous root that comes in when the force (1) is used;[2] the root $a(1 - i)$ gives a non-radiating orbit. Any orbit of the whole set

$$z = C_1\,e^{-a(1-i)t} + C_2\,e^{a(1-i)t} \tag{7}$$

is also non-radiating.

The orbits (4) have been described as non-radiating because the force (1), being normal to the path, does no work. It is only in this sense that they are free from radiation: the equation of energy holds, the sum of the kinetic and potential energies is constant. (As the motions are supposed to be small, quasi-stationary, relativity effects have been ignored.) From the point of view of electro-magnetic theory the rate of radiation varies as the square of the acceleration and can never vanish in accelerated motion; and there is a coördinate radiation of momentum. The relation between the two rates of radiation is shown by the equation

$$\frac{d}{dt}\left(\mathbf{v}\cdot\frac{d\mathbf{v}}{dt}\right) = \frac{d\mathbf{v}}{dt}\cdot\frac{d\mathbf{v}}{dt} + \mathbf{v}\cdot\frac{d^2\mathbf{v}}{dt^2}$$

The total radiation is the same according to both points of view whenever estimated between two instants for which $\mathbf{v}.d\mathbf{v}/dt$ has the same values; but the instantaneous rates cannot be indentically equal except for orbits in which $\mathbf{v}.d\mathbf{v}/dt$ is always the same constant independent of the time. I have discussed elsewhere the effect of replacing the law (1) by its alternative in the case of the rectilinear oscillation,[3]—which then ceases to be an oscillation.

From the point of view of relativity the rate of radiation of energy (and momentum) has been treated by several authors and has been shown to vary with the square of the generalized curvature of the path when regarded as a space-time locus in four dimensions.[4] For any real motion, where the velocity is less than that of light, the radiation must be positive and radiationless orbits, other than straight lines uniformly described, are impossible. If, however, velocities greater than light were possible and if the formula for the rate of radiation still held, the orbits would be those for which the curvature vector was a minimum line. Although this state of affairs may have no present

physical interest, it does have a purely mathematical interest, and I shall therefore determine also these orbits.

The expression which must be integrated is [4]

$$\frac{d\mathbf{v}}{dt} \cdot \frac{d\mathbf{v}}{dt} = \left(\mathbf{v} \times \frac{d\mathbf{v}}{dt}\right) \cdot \left(\mathbf{v} \times \frac{d\mathbf{v}}{dt}\right), \tag{8}$$

provided the units are so chosen that the velocity of light is 1. Let R be the radius of curvature of the orbit (in ordinary space). The tangential and normal resolution of acceleration throws (8) into

$$\left(\frac{dv}{dt}\right)^2 + \frac{v^4}{R^2} = \frac{v^6}{R^2} \quad \text{or} \quad \left(\frac{dv}{ds}\right)^2 = \frac{v^4 - v^2}{R^2}.$$

Hence

$$v = \sec \int \frac{ds}{R}. \tag{9}$$

If then any space curve be given intrinsically by the equation $R = f(s)$, equation (9) determines the velocity at which the curve must be traced if there is to be no radiation as estimated by the usual electro-magnetic formula.

[1] See H. A. Lorentz, *Theory of Electrons*, Art. 37, B. G. Teubner, Leipzig, 1916.
[2] See M. Planck, *Theorie der Wärmestrahlung*, p. 110, J. A. Barth, Leipzig, 1906.
[3] Wilson, E. B., *Boston, Proc. Amer. Acad. Arts Sci.*, 50, 1914, (105–128).
[4] See Wilson and Lewis, *Ibid.*, 48, 1912, (387–507), especially p. 481.

THE EFFECT UPON THE ATOM OF THE PASSAGE OF AN ALPHA RAY THROUGH IT[1]

By R. A. Millikan, V. H. Gottschalk and M. J. Kelly

Ryerson Physical Laboratory, University of Chicago

Read before the Academy, November 11, 1919

In 1910, by catching at the instant of ionization the positive residues of atoms ionized by X-rays, and by beta and gamma rays of radium, it was conclusively shown[2] that the act of ionization by these agencies uniformly consists in the detachment of a single negative electron from a neutral atom.

The method consisted in balancing the force of gravity acting upon a minute oil-drop by a strong vertical electrical field, holding the oil-drop under observation in a telescope with the aid of a powerful beam of light,

passing a sharply limited beam of X-rays, beta rays or γ rays immediately underneath the drop, catching upon the drop the positive ion, formed by the ionization of a neutral molecule by the rays under investigation, and finally measuring the charge communicated to the drop by the advent of the ion upon it through observing the speed imparted to the drop by its new increment in charge.

Just before the war, Millikan attacked the more difficult and the more interesting problem of catching by the same general method, the ions formed by the passage of an alpha particle through an atom, expecting in this case to find that this relatively huge and powerful ionizing agent would often detach more than one negative electron from a single atom. When he was called to other duties by the war, the experimental work already begun was continued and completed by Gottschalk and Kelly. The results are as follows:

1. Alpha rays have been shot through atoms of the most diverse sorts (H, C, O, N, Cl, I, Hg) and of atomic weights from 1 to 200, without bringing to light in any case evidence of the formation of multiply-valent ions.

2. Twenty-nine hundred ions formed by the passage of α rays through neutral molecules have been caught on oil drops at the instant of ionization and the charges carried by each of these ions individually measured. Of these 2900 captures, 5 might possibly have corresponded to double charges, though even these were in all probability due to the nearly simultaneous advent upon the drop of two singly charged ions.

3. In no single case has an α particle been observed to form an ion carrying three or more charges, even though mercury, from which octivalent ions had been expected, was one of the substances tested.

4. *Alpha ray ionization consists, then at least 99 times out of a 100, in the case of all the gases and vapors studied, in the detachment of a single negative electron from a neutral molecule.*

[1] A detailed report of these experiments will shortly be published in the *Physical Review*.
[2] Millikan, R. A., and Fletcher, H., *London Phil. Mag.*, (Ser. 6) 21, 1911, (753).

ON THE EMBRYOLOGICAL BASIS OF HUMAN MORTALITY[1]

By Raymond Pearl

Department of Biometry and Vital Statistics, Johns Hopkins University

Read before the Academy, November 11, 1919

1. In order to get a clearer idea of the underlying biological factors in human mortality I have rearranged the 'causes of death' listed in the International Classification of the Causes of Death, which is the code used generally by vital statisticians, into a new classification on a biological basis. It is not possible with our present statistical material to make a completely and precisely logical classification, but I have endeavored to come as close to it as is possible. The underlying idea of this new classification is, as the first operation, to group all causes of death under the heads of the several organ systems of the body, the functional breakdown of which is the immediate or predominant cause of the cessation of life. All except a few of the statistically recognized causes of death in the International Classification can be assigned places in such a biologically grouped list. It has a sound logical foundation in the fact that, biologically considered, death results because some organ system, or group of organ systems, fails to continue its functions. Practically, the plan involves the reassignment of all of the several causes of death now grouped by vital statisticians under heading 'I. General diseases.' It also involves the re-distributing of causes of death now listed under the puerperal state, malformations, early infancy, and certain of those under external causes.

The headings finally decided upon for the new classification are as follows:

I. Circulatory system, blood, and blood-forming organs
II. Respiratory system
III. Primary and secondary sex organs
IV. Kidneys and related excretory organs
V. Skeletal and muscular systems
VI. Alimentary tract and associated organs concerned in metabolism
VII. Nervous system and sense organs
VIII. Skin
IX. Endocrinal system
X. All other causes of death

It should be emphasized that the underlying idea of this rearrangement of the causes of death is to put all those lethal entities together which bring about death because of the functional organic breakdown of the same general organ system. The cause of this functional breakdown may be anything whatever in the range of pathology. It may be due to bacterial infection; it may be due to trophic disturbances; it may be due to mechanical disturbances which prevent the continuation of normal function; or to any other cause whatsoever. In other words, the basis of the present classification is not that of pathological causation, but it is rather that of organological breakdown. We are now looking at the

TABLE 1

SHOWING THE RELATIVE IMPORTANCE OF DIFFERENT ORGAN SYSTEMS IN HUMAN MORTALITY

GROUP NUMBER	ORGAN SYSTEM	DEATH RATES PER 100,000			
		Registration area, U. S. A.		England and Wales, 1914	São Paulo, 1917
		1906–10	1901–05		
II	Respiratory system	395.7	460.5	420.2	417.5
VI	Alimentary tract and associated organs	334.9	340.4	274.1	613.8
I	Circulatory system, blood	209.8	196.8	208.6	254.8
VII	Nervous system and sense organs	175.6	192.9	151.9	124.3
IV	Kidneys and related excretory organs	107.2	107.4	19.4	83.4
III	Primary and secondary sex organs	88.1	77.4	95.4	103.2
V	Skeletal and muscular system	12.6	13.7	18.2	6.8
VIII	Skin	10.1	13.3	12.0	7.9
IX	Endocrinal system	1.5	1.2	1.9	1.1
	Total death rate classifiable on a biological basis	1335.5	1403.6	1231.7	1612.8
X	All other causes of death	171.3	211.9	141.4	109.8

question of death from the standpoint of the pure biologist, who concerns himself not with what causes a cessation of function, but rather with what part of the organism ceases of function, and therefore causes death.

2. In table 1 the death rates per 100,000 are arranged in descending order of importance (for the United States Registration Area 1906–10) by organ systems. Four sets of data are used: (a) the United States Registration Area for the five years, 1906–10 inclusive; (b) the same, 1901–05; (c) England and Wales, 1914; and (d) São Paulo, Brazil, 1917.

3. The data show that in the United States, during the decade covered, more deaths resulted from the breakdown of the respiratory system than from the failure of any other organ system of the body. The same thing

is true of England and Wales. In Sao Paulo the alimentary tract takes first position, with the respiratory system a rather close second. The tremendous death rate in Sao Paulo chargeable to the alimentary tract is chiefly due to the relatively enormous number of deaths of infants under two from diarrhea and enteritis. Nothing approaching such a rate for this category as Sao Paulo shows is known in this country or England.

In all three localities studied the respiratory system and the alimentary tract together account for rather more than half of all the deaths biologically classifiable. These are the two organ systems which, while physically internal, come in contact directly at their surfaces with environmental entities (water, food, and air) with all their bacterial contamination. The only other organ system directly exposed to the environment is the skin. The alimentary canal and the lungs are, of course, in effect invaginated *surfaces* of the body. The mucous membranes which line them are far less resistant to environmental stresses, both physical and chemical, than is the skin with its protecting layers of stratified epithelium.

The organs concerned with the blood and its circulation stand third in importance in the mortality list. Biologically the blood, through its immunological mechanism constitutes the second line of defense which the body has against noxious invaders. The first line is the resistance of the outer cells of the skin and the lining epithelium of alimentary tract, lungs, and sexual and excretory organs. When invading organisms pass or break down these first two lines of defense the battle is then with the home guard, the cells of the organ systems which, like the industrial workers of a commonwealth, keep the body going as a whole functioning mechanism. Naturally it would be expected that the casualties would be far heavier in the first two defense lines (respiratory and alimentary systems, and blood and circulation) than in the home guard. Death rates when biologically classified bear out this expectation.

It is at first thought somewhat surprising that the breakdown of the nervous system is responsible for more deaths than that of the excretory system. When one bears in mind, however, the relative complexity of the two pieces of machinery, it is perceived that the relative position of the two in responsibility for mortality is what might be reasonably be expected.

In the United States the kidneys and related excretory organs are responsible for more deaths than the sex organs. This relation is reversed in England and Wales and in Sao Paulo. The difference is mainly

due, in the case of England, to two factors, premature birth and cancer. In Sao Paulo it is due to premature birth and syphilis.

In a broad sense the efforts of public health and hygiene have been directed against the affections comprised in the first two items in the table, respiratory system and alimentary tract. The figures in the first two columns for the two five year periods in the United States indicate roughly the rate of progress such measures are making, looking at the matter from a broad biological standpoint. In reference to the respi-

TABLE 2

SHOWING THE RELATIVE INFLUENCE OF THE PRIMARY GERM LAYERS IN HUMAN MORTALITY

(Items 64 and 65 charged to ectoderm)

LOCALITY	DEATH RATE PER 100,000 DUE TO FUNCTIONAL BREAKDOWN OF ORGANS EMBRYOLOGICALLY DEVELOPING FROM					
	Ectoderm	Per cent	Meso-derm	Per cent	Endo-derm	Per cent
United States Registration Area, 1906–10	191.1	14.3	425.2	31.8	719.6	53.9
United States Registration Area, 1901–05	210.6	15.0	407.1	29.0	786.2	56.0
England and Wales, 1914	177.1	14.4	374.0	30.3	681.5	55.3
Sao Paulo, 1917	134.9	8.4	468.0	29.0	1009.9	62.6

TABLE 3

SHOWING THE RELATIVE INFLUENCE OF THE PRIMARY GERM LAYERS IN HUMAN MORTALITY

(Items 64 and 65 charged to mesoderm)

LOCALITY	DEATH RATE PER 100,000 DUE TO FUNCTIONAL BREAKDOWN OF ORGANS EMBRYOLOGICALLY DEVELOPING FROM					
	Ectoderm	Per cent	Meso-derm	Per cent	Endo-derm	Per cent
United States Registration Area, 1906–10	116.9	8.7	499.4	37.4	719.6	53.9
United States Registration Area, 1901–05	137.3	9.8	480.4	34.2	786.2	56.0
England and Wales, 1914	107.9	6.7	443.2	36.0	681.5	55.3
Sao Paulo, 1917	101.3	6.3	501.6	31.1	1009.9	62.6

ratory system there was a decline of 14% in the death rate between the two periods. This is substantial. It is practically all accounted for in phthisis, lobar pneumonia, and bronchitis. For the alimentary tract the case is not so good—indeed, far worse. Between the two periods the death rate from this cause group fell only 1.8%.

4. The next step in the investigation was to arrange all the organologically classifiable death rates under the primary germ layers (ectoderm, mesoderm, and endoderm) from which the organs concerned developed embryologically. The results are set forth in tables 2 and 3, and in

figure 1. Tables 2 and 3 give upper and lower limiting values to the death rates chargeable to ectoderm and mesoderm. The difference between the two depends upon the placing of deaths due to cerebral hemorrhage and apoplexy, and to 'softening of the brain'. The discussion of the embryological and pathological problems involved cannot be given here by reason of lack of space. The complete paper must be consulted.

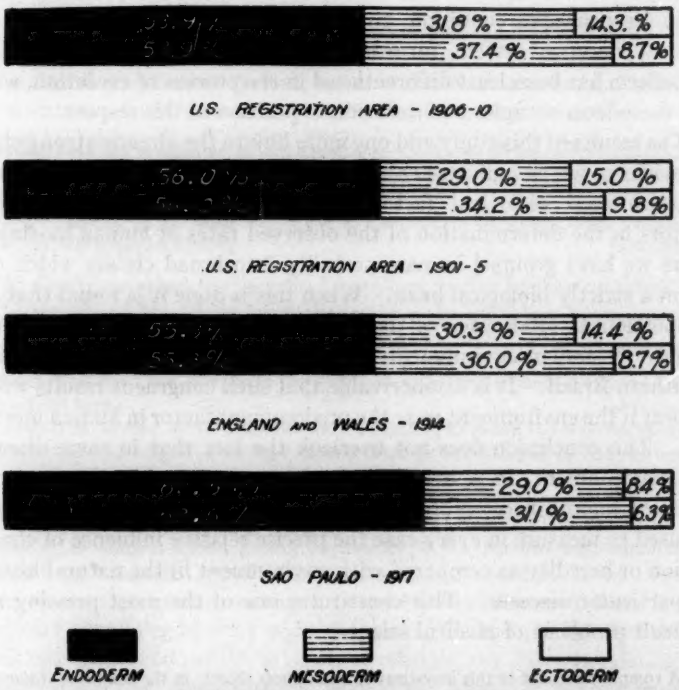

FIG. 1. DIAGRAM, SHOWING THE PERCENTAGES OF BIOLOGICALLY CLASSIFIABLE HUMAN MORTALITY RESULTING FROM BREAKDOWN OF ORGANS DEVELOPING FROM THE DIFFERENT GERM LAYERS

Upper bar of pair gives upper limit of mortality chargeable to ectoderm; lower bar gives lower limit of mortality chargeable to ectoderm.

The data of tables 2 and 3 are shown graphically in percentage form in figure 1.

The final results lead to a generalization of considerable interest and significance to the evolutionist. The figures show that in man, the highest product of organic evolution, about 57% of all the biologically classifiable deaths result from a breakdown and failure further to function

of organs arising from the endoderm in their embryological development while but from 8 to 13% can be regarded as a result of breakdown of organ systems arising from the ectoderm. The remaining 30 to 35% of the mortality results from failure of mesodermic organs. Taking a general view of comparative anatomy and embryology it is evident that in the evolutionary history through which man and the higher vertebrates have passed it is the ectoderm which has been most widely differentiated from its primitive condition, to the validity of which statement the central nervous system furnishes the most potent evidence. The endoderm has been least differentiated in the process of evolution, while the mesoderm occupies an intermediate position in this respect.

The results of this study add one more link to the already strong chain of evidence which indicates the highly important part played by innate constitutional biological factors as contrasted with environmental factors in the determination of the observed rates of human mortality. Here we have grouped human mortality into broad classes which rest upon a strictly biological basis. When this is done it is found that the proportionate subdivision of the mortality is strikingly similar in such widely dissimilar environments as the United States, England, and Southern Brazil. It is inconceivable that such congruent results would appear if the environment were the predominant factor in human mortality. This conclusion does not overlook the fact that in some diseases the environment, in a broad sense, is unquestionably the factor of greatest importance. Nor does it imply that every effort should not be used to measure in every case the precise relative influence of constitution or heredity as compared with environment in the natural history of particular diseases. This constitutes one of the most pressing and difficult problems of medical science.

[1] A complete account of this investigation will appear shortly in the *American Naturalist*. That paper must be consulted for detailed discussion of the intricate pathological and embryological points involved in the statistical treatment of the data in this investigation.

NATIONAL RESEARCH COUNCIL

EXTRACTS FROM THE MINUTES OF THE MEETING OF THE INTERIM COMMITTEE

AT THE OFFICE OF THE NATIONAL RESEARCH COUNCIL, AUGUST 19, 1919, AT 10.00 A.M.

Mr. Angell in the chair.

Present: Messrs. Angell, Barrows, Howe, Ransome, Wood, and by invitation Mr. J. H. J. Yule.

The Chairman presented the resignation of Dr. W. F. Durand as Chairman of the Patent Office Committee of the National Research Council.

Moved: That the resignation of Mr. Durand as Chairman of the Patent Office Committee be accepted with an expression of thanks from the Council for the work which he has done as chairman of this committee. (*Adopted.*)

Moved: That Mr. L. H. Baekeland be elected Chairman of the Patent Office Committee of the National Research Council. (*Adopted.*)

The Chairman asked the Interim Committee to give further consideration to the request of Mr. Wade for authority to make purchases against appropriations and allotments already provided for.

Moved: That the Chief Clerk with the Bursar be authorized to make purchases against appropriated funds. (*Adopted.*)

Moved: That in view of the inability of Mr. Leuschner to return to his duties as Secretary, Mr. Wood be designated Acting Secretary until further action of the Committee. (*Adopted.*)

The Chairman asked informal consideration as to the advisability of the Chairman sending a letter, giving an outline of the present organization of the National Research Council without personnel, to the heads of the Departments of the Government, stating that the National Research Council had now, after completing its work under the war organization, reorganized under peace conditions and would be glad to consider any matters relating to research which they might desire to have advice upon. It was the sense of the meeting that the Chairman should prepare such a letter and send it to the heads of the Departments of the Government.

The meeting adjourned at 11.25 a.m.

PAUL BROCKETT, *Assistant Secretary.*

EXTRACTS FROM THE MINUTES OF THE MEETING OF THE INTERIM COMMITTEE

AT THE OFFICE OF THE NATIONAL RESEARCH COUNCIL, OCTOBER 2, 1919, AT 9.15 A.M.

Mr. Angell in the chair.

Present: Messrs. Angell, Bancroft, Barrows, H. E. Howe, Kellogg, McClung, Mathews, Mendenhall, Ransome, Wood, Yerkes.

The minutes of the meeting of the Interim Committee of the National Research Council of August 19 were approved as circulated.

The Chairman, as chairman of the Finance Committee, rendered an informal report of a meeting of that committee held in New York on September 9.

Mr. Howe, Chairman of the Committee on Organization and Budget, reported that, after consultation with the Treasurer, he had taken out insurance to the amount of $5000 on that portion of the property contained in the offices of the Council which belongs to the National Research Council.

Moved: That the action taken with reference to insurance be approved. (*Adopted.*)

Mr. Kellogg, Chairman of the Publicity Committee, presented an informal report of the activities of that committee.

On recommendation of Mr. Yerkes, Chairman of the Committee on Publication, it was

Moved: That the Committee on Publication be authorized to publish Bulletins of the National Research Council in appropriate editions. (*Adopted.*)

Mr. Yerkes presented an informal report of the Committee on Publication concerning difficulties which had arisen in the publication of Bulletins of the Council, such as questions of postal rates, economy of printing, and the relation of the Bulletins of the Council to the PROCEEDINGS of the National Academy of Sciences. Action regarding the form of publication was postponed until the next meeting of the Executive Board.

The Vice Chairman of the Division of Industrial Relations made an informal report of the progress of the Division in its various activities.

On account of certain objections offered by one of the Divisions of Science and Technology to the change of name to Division of Industrial Research, the matter was again brought up for consideration. The Vice Chairman of the Division made a clarifying statement setting forth the actual functions of the Division. It was the sense of the meeting that the recommendation already made to the National Academy of Sciences, that the name be changed to Division of Industrial Research should be allowed to stand.

On recommendation of the Vice Chairman of the Division it was

Moved: That the exhibit to be put in by the Western Electric Company, all expenses of which they are to defray, be authorized. (*Adopted.*)

The Chairman of the Research Information Service drew attention to a recommendation made by the Committee on International Catalogue of Scientific Literature, which provides that the catalogue be changed to simply an author's list of publications arranged under the proper division of science, but not subdivided further, and that this list be published once in five years. In view of the thought given by the Council to the matter of abstracts and

bibliographies, and of its plans for the improvement of the status of scientific publications, and since it will be necessary for the Council to make individual response to this recommendation through the Foreign Secretary of the National Academy, it was

Moved: That a committee representing the several divisions of the Research Council be appointed by the Chairman, who shall serve as Chairman of the committee, to consider existing provisions for the abstracting and indexing of scientific literature in their relations to the proposed modification of the International Catalogue of Scientific Literature, and to formulate a plan for improving methods of preparing and issuing scientific abstracts and bibliographic lists. (*Adopted.*)

The Chairman of the Research Information Service requested reconsideration of the action of the Executive Board at its meeting on June 24, 1919, which disapproved the resolution of the Research Information Service requesting authorization to charge for its services for those exceptional cases which involve considerable time and labor in the search for the necessary information, or in special investigations.

Moved: That the Interim Committee ask the Executive Board to reconsider its action of June 24, relative to charges to be made by the Research Information Service for services in exceptional cases. (*Adopted.*)

The Chairman of the Division of Chemistry and Chemical Technology reported that in accordance with action of the Interim Committee on June 3, 1919, three trustees had been appointed for the business control of the publication of critical tables of physical and chemical constants, namely, Mr. Hugh K. Moore, appointed by the Chairman of the National Research Council; Mr. Julius Stieglitz, appointed by the President of the American Chemical Society; and Mr. A. L. Day, appointed by the President of the American Physical Society, who has not yet signified his acceptance of the appointment. A campaign has been proposed for securing the sum of $100,000 to carry on the work of publication, for the initial expenses of which a loan of $1500 has been requested by the Trustees.

Moved: That the Chairman of the Division of Physical Sciences and of the Division of Chemistry and Chemical Technology be authorized to transfer to this fund $500 each, with the understanding that this money shall be paid back to the account of these Divisions out of the subscriptions as soon as subscriptions come in. (*Adopted.*)

The Chairman of the Division reported that the Subcommittee on Ceramic Research has voted, with the approval of the Division, to secure funds for fellowships in ceramic research.

Moved: That the Committee on Ceramic Research be authorized to solicit funds for fellowships in ceramic research, as outlined in the minutes of the meeting of the Committee on Ceramic Research held on June 23, 1919. (*Adopted.*)

The Chairman of the Division of Educational Relations presented for the information of the Interim Committee a plan for a survey of research conditions in American Colleges and Universities.

The Chairman of the Division reported that a request for the appointment of Mr. Herbert Hoover as a Member at large of the Division of Educational Relations has been filed with the Secretary.

Moved: That the continuation of the appointment of Mr. Alfred D. Flinn, Secretary of the Engineering Foundation, as Assistant Secretary, by reciprocal arrangement, of the National Research Council, be confirmed. (*Adopted.*)

The Chairman presented a letter addressed to the Council by Mr. W. E. Mosher for the Commission of Congress dealing with the reclassification of salaries of scientific men, requesting the cooperation of the Council in securing data relative to salaries and duties of scientific men in universities and industrial institutions throughout the country.

Moved: That Mr. Mosher be invited to confer with Chairman of Divisions of the Council in regard to the information they desire. (*Adopted.*)

Moved: That the Chairman be authorized to appoint a Committee on By-Laws of the Council. (*Adopted.*)

The Chairman thereupon appointed Messrs. Angell, Mendenhall, and Wood.

On recommendation of the Acting Secretary it was

Moved: That the name, Editorial Committee, be changed to Committee on Publication. (*Adopted.*)

The Acting Secretary presented a letter of appreciation from Mr. Whitman Cross containing a request that the period of his services as Treasurer of the Council be shown as one year and four months instead of two and one-half years, in the resolutions of the Executive Board of August 12, 1919.

(*Approved.*)

The Chairman reported that Mr. Vernon Kellogg had consented to assume conjointly the work of the secretarial office and the chairmanship of the Division of Educational Relations, Mr. Wood to continue in his present position.

The Chairman asked judgment of a plan proposed by Mr. Merriam for meetings at frequent intervals during several weeks to come in order that the Chairmen may keep closely in touch with the work of other Divisions. The plan was approved and the arrangement for such meetings left to the Chairman.

Regarding the regular meetings of the Interim Committee it was

Moved: That for the present meetings of the Interim Committee be held weekly on Tuesday at nine o'clock. (*Adopted.*)

The meeting adjourned at 12.45 p.m. until 9.00 a.m., Tuesday, October 7.

HARRY O. WOOD, *Acting Secretary.*

EXTRACTS FROM THE MINUTES OF THE MEETING OF THE INTERIM COMMITTEE

AT THE OFFICE OF THE NATIONAL RESEARCH COUNCIL, OCTOBER 7, 1919 AT 9.00 A.M.

Mr. Angell in the chair.

Present: Messrs. Angell, Bancroft, Barrows, Brockett, Christian, Kellogg, McClung, Mathews, Mendenhall, Ransome, Wood, and Yerkes.

Mr. Yerkes, Chairman of the Committee on Publication, reported that a conference had been arranged with Mr. Pearl for the afternoon of October 7, and asked the advice of members of the Interim Committee regarding questions to be discussed at the conference. This led to a discussion of the matter of publications at considerable length.

Mr. Kellogg, Chairman of the Publicity Committee, reported that arrangements had been made for the distribution of any information concerning the activities of the Council of sufficient news value to be published in newspapers. The Chairman of the Committee read a letter to be sent to Chairmen of Divisions requesting that all matter arising in the various Divisions which may be used in this connection be submitted to the Publicity Committee for distribution.

The Acting Chairman of the Division of Geology and Geography reported that the recommendations contained in the resolution of June 10, 1919, relative to coördination in field methods of map-making agencies of the Government had already been attended to by the Division of Geology and Geography, and that a complete report upon this would be furnished the Government Division after its organization has been completed.

The Chairman of the Council extended the welcome of the Interim Committee to Dr. Christian, and expressed the pleasure of its members at his arrival in Washington to participate in the activities of the Council.

The Chairman of the Council reported that six members of the Section on Psychology had already been selected and that a meeting of these is to be held in Washington on October 20. He reported further that six members of the Section on Anthropology had also been selected and it was hoped that a joint meeting of the two sections could be held on October 20.

The meeting adjourned at 10.45 a.m.

HARRY O. WOOD, *Acting Secretary.*

EXTRACTS FROM THE MINUTES OF THE MEETING OF THE EXECUTIVE BOARD

AT THE OFFICE OF THE NATIONAL RESEARCH COUNCIL, OCTOBER 14, 1919, AT 9.30 A.M.

Mr. Angell in the chair.

Present: Messrs. Abbot, C. A. Adams, Angell, Bancroft, Christian, Flinn, Johnston, Kellogg, McClung, Manning, Mathews, Mendenhall, Pearl, Pupin, Ransome, Townsend, Walcott, Woodward, Yerkes, and by invitation, Barrows, Brockett, Wood.

The minutes of the meetings of the Executive Board of the National Research Council on August 12 and of the Interim Committee on October 2 were approved as circulated with certain slight textual corrections.

Moved: That Mr. Vernon Kellogg, Chairman of the Division of Educational Relations, be appointed to serve also as Secretary of the National Research Council, to date from October 1, 1919. (*Adopted.*)

Moved: That Mr. Harry O. Wood, Acting Secretary of the National Research Council, be appointed Assistant Secretary. (*Adopted.*)

Confirming action taken by the Division of Engineering at its meeting on September 29, 1919, it was

Moved: That the Executive Board approve the appointment of Mr. Comfort A. Adams as Chairman of the Division of Engineering. (*Adopted.*)

Moved: That Mr. Henry M. Howe be appointed Honorary Chairman of the Division of Engineering for one year. (*Adopted.*)

Moved: That Mr. Galen H. Clevenger be appointed Vice-Chairman of the Division of Engineering without salary. (*Adopted.*)

The Treasurer presented a financial report for the period ending September 30, 1919.

The Chairman reported that Mr. Abraham Flexner, Secretary of the General Education Board, writing under date of September 6, 1919, had transmitted to him the following action of that Board taken on September 5, 1919:

Resolved, That, upon the request of the National Research Council under date of August 12, 1919, the sum of Ten Thousand Dollars ($10,000), or so much thereof as may be needed, be, and it hereby is, appropriated to the National Research Council, to enable the Division of Educational Relations to conduct certain educational investigations outlined in Dr. Angell's memorandum during the year ending September 30, 1920.

In view of the action of the General Education Board be it

Resolved, That the National Research Council accept with cordial appreciation the generous gift of the General Education Board of Ten Thousand Dollars ($10,000) for the special work of the Division of Educational Relations of the Council in making an investigation of the conditions affecting research in American educational institutions; and that the Council will exercise every effort to make the use of this gift yield results of advantage to American science and education. (*Approved.*)

Mr. Kellogg, Chairman of the Committee on Publicity, reported various steps taken to make the purposes and activities of the Council more widely known to the general public and the scientific men of the country through the newspaper press and technical journals.

The Chairman of the Division of Physicial Sciences, in response to a request from the Division of States Relations

Moved: That Professor Ernest Merritt be appointed to represent the Division of Physical Sciences in the Division of States Relations. (*Adopted.*)

The Chairman of the Division of Engineering reported the appointment of a committee for the investigation of high speed steel, of which Mr. J. V. Emmons has been named Chairman.

Moved: That the appointment of a committee for the investigation of high speed steel be approved. (*Adopted.*)

The Chairman of the Division of Medical Sciences reported that Professor Graham Lusk had been nominated by the Society of Biological Chemists as its representative in the Division of Medical Sciences.

Moved: That the nomination of Professor Graham Lusk be received and transmitted to the President of the National Academy of Sciences with the recommendation that he be appointed a member of the National Research Council and assigned to the Division of Medical Sciences. (*Adopted.*)

The Chairman of the Division of Biology and Agriculture presented for reconsideration a request that the National Research Council in raising funds for the support of Marine Biological Laboratory at Woods Hole. He emphasized the importance of the work carried on at this laboratory, which is handicapped at the present time by lack of funds. He asked that the Council take action.

After a short discussion consideration of the matter was postponed until the afternoon session.

The following nomination was presented by the Chairman of the Division for member at large of the Division of Educational Relations:

Mr. Herbert Hoover, Ex-United States Food Administrator; Trustee, Leland Stanford, Jr., University.

Moved: That the nomination of Mr. Herbert Hoover be approved with the recommendation to the President of the National Academy of Sciences that he be appointed a member of the National Research Council and assigned to the Division of Educational Relations. (*Adopted.*)

Upon recommendation of the Chairman of the Division it was

Moved: That Col. Robert I. Rees be appointed a representative from the Committee on Education and Special Training of the War Department as a member of the Division of Educational Relations to fill a vacancy in that position. (*Adopted.*)

The Chairman of the Council announced that the Division of Anthropology and Psychology is in process of organization, and that a meeting of the Division will be held on October 20.

The Chairman of the Research Information Service presented the following matters:

In response to a request from the Division of States Relations it was

Moved: That Mr. Edwin F. Gay be appointed to represent the Research Information Service in the Division of States Relations. (*Adopted.*)

The following nomination was presented by the Chairman of the Division for membership in that Division:

Mr. Frank Burke, Chief of the Bureau of Investigation of the Department of Justice.

Moved: That the name of Mr. Frank Burke, Chief of the Bureau of Investigation, as representative of the Department of Justice, be presented to the President of the United States in accordance with Article V, Section 3, of the Organization of the National Research Council, for designation by him for service with the National Research Council on the Research Information Service. (*Adopted.*)

The Chairman of the Division presented the following nominations for members at large of the Research Information Service:

Clement W. Andrews, Librarian, The John Crerar Library, Chicago, Illinois.

Milton C. Whitaker, Consulting Chemical Engineer, New York City.

Arthur D. Little, President, Arthur D. Little, Inc., Industrial Research Laboratories, 30 Charles River Road, Cambridge, Mass.

Fuller R. Callaway, LaGrange, Ga., Cotton Manufacturer.

F. S. Terry, Mgr., National Lamp Works of General Electric Co., Nela Park, Cleveland, Ohio.

W. R. De Field, President, W. R. De Field & Co., Chicago, Ill.

Charles F. Kettering, President, The Dayton Engineering Laboratories Co., Dayton, Ohio.

Moved: That the nominations of Messrs. Clement W. Andrews, Milton C. Whitaker, Arthur D. Little, Fuller R. Callaway, F. S. Terry, Charles F. Kettering, and W. R. DeField be approved with the recommendation to the President of the National Academy that they be appointed members of the National Research Council and assigned to the Research Information Service. (*Adopted.*)

The Chairman of the Research Information Service called attention to action taken by the Interim Committee at its meeting on October 2, 1919, recommending to the Executive Board reconsideration of its action on June 24, 1919, relative to the making of charges by the Research Information Service for services in exceptional cases. It appeared to be the sense of the meeting that such charges would be proper, but in view of the absence of a member of this Board, who had previously opposed the proposal, it was

Moved: That the matter be referred to the Interim Committee with power, and that an effort be made to consult absent members as to any grounds of opposition. (*Adopted.*)

The Chairman of the Council reported that, with the return of Mr. Walcott to the city, action is being taken in the organization of the Government Division, and it is hoped that it will be completed within a short time.

The Executive Secretary of the Division of States Relations reported the following nominations from the Divisions named to serve as members of the Division of States Relations:

For the Division of Industrial Relations: Mr. H. E. Howe.

For the Division of Educational Relations: Mr. Vernon Kellogg.

For the Division of Engineering: Mr. S. W. Stratton.

For the Division of Chemistry and Chemical Technology: Mr. W. D. Bancroft.

For the Division of Geology and Geography: Mr. E. B. Mathews.

Moved: That the nominations of Messrs. H. E. Howe, Vernon Kellogg, S. W. Stratton, W. D. Bancroft, and E. B. Mathews as members of the Division of States Relations be approved. (*Adopted.*)

The Executive Secretary of the Division of States Relations also reported the nomination from the Society of American Foresters of Mr. Hugh P. Baker to serve on the Division of States Relations.

Moved: That the nomination of Mr. Hugh P. Baker be approved, with the recommendation to the President of the National Academy of Sciences that he be appointed a member of the National Research Council and assigned to the Division of States Relations. (*Adopted.*)

The Executive Board took recess at 12.30 to meet again at 2.00 p.m.

The Executive Board resumed its meeting at 2.20 p.m.

The Chairman made reference to the matter of quorum and stated that consideration of this was referred to the Committee on By-Laws.

At the suggestion of the Chairman the legal relations of the National Research Council to the National Academy of Sciences were discussed, and it was

Moved: That the Chairman of the Council be authorized and requested to pursue this subject farther with a view to clarifying the status of the Council. (*Adopted.*)

The Chairman of the Council raised the question of policy in regard to research in the industries, especially as this relates to overlapping of interests in the various Divisions and, in particular, in the Divisions of Industrial Relations and of Engineering. A discussion at length followed.

Moved: That a committee of five on General Policy and Solicitation of Funds, of which the Chairman of the Council shall act as Chairman, be appointed. (*Adopted.*)

Appointed: Messrs. Angell (Chairman), C. A. Adams, H. E. Howe, Kellogg, and Walcott.

The question of securing financial aid for the Marine Biological Laboratory, which had been presented at the forenoon session, was brought up for further consideration.

Moved: That the matter of securing financial assistance for the Marine Biological Laboratory be referred for recommendation to the Committee on General Policy and Solicitation of Funds. (*Adopted.*)

The Chairman requested an expression of opinion by the Board concerning the policy of the Council in reference to matters of congressional legislation, with special reference to two bills at present pending,—one providing for a tax on scientific instruments, and the other the removal of exemption which has hitherto been enjoyed by educational institutions. It was the sense of the meeting that the Council should take no action.

The Chairman of the Council presented a recommendation submitted by Mr. Dunn, as Chairman of a committee appointed by the Executive Board on June 3, 1919, to provide for the continuation of retired Chairmen of the Council as members of the Executive Board.

Moved: That the Executive Board recommend to the Council of the National Academy of Sciences that Article IV, Section 2, of the Organization of the National Research Council be amended by the addition of the following sentence: "Chairmen, on their retirement, shall continue for three years as members of the Executive Board." (*Adopted.*)

The Chairman presented a recommendation from Mr. Alfred D. Flinn, that the action of the Interim Committee on October 2, 1919, providing for the continuation of the appointment of Mr. Flinn as Assistant Secretary of the National Research Council be amended to omit the name of Mr. Flinn, making reference only to the Secretary of the Engineering Foundation.

(*Approved.*)

On motion of the Treasurer various small items of appropriation and accounting were approved.

The Chairman stated that in the allotment of funds for the current year certain expenses had not been provided for, making it necessary for the Executive Board to appropriate small sums of money under various headings to cover such unauthorized items. He suggested that this matter should be referred to the Committee on Organization and Budget.

Moved: That the Committee on Organization and Budget be requested to make a redistribution of allotments of general expense temporarily and to prepare a motion to authorize action in future. (*Adopted.*)

The matter of the publication of minutes of Divisions, committees, etc., was discussed, and it was

Moved: That nothing be published by the National Research Council or any of its Divisions without the sanction of the Committee on Publication. (*Adopted.*)

The meeting adjourned at 4.20 p. m. to meet again at 9.30 a. m., November 18.

HARRY O. WOOD, *Assistant Secretary.*

INDEX

ABBOT, C. G. Simultaneous Measurements of Solar Constant of Radiation at Mount Wilson, California and Calama, Chile, 383.
——. Rotating Projectiles from Smooth-Bore Guns, 386.
——. Measuring Speed of Projectiles in Flight, 388.
Abelian varieties (Lefschetz), 296.
Acoustical impedance (Webster), 275.
Acoustical measurements (Webster), 173.
ADAMS, W. S., and JOY, A. H. Motions in Space of some Stars of High Radial Velocity, 239.
—— and STRÖMBERG, G. Spectroscopic Method for determining Parallaxes of the Brighter Stars, 228.
ADLER, L., see MacInnes, D. A., 160.
Adsorption by charcoal (Lemon and Blodgett), 289; (Lemon), 291.
Alpha rays (Millikan *et al.*), 591.
Annual Meeting of the Academy, 479.
Aphelia in the solar system (Perrine), 371.
Archaeology of the Southwest (Nelson), 114.
AREY, L. B. and CROZIER, W. J. Nervous Organization of a Nudibranch, 498.
AREY, L. B., see Crozier, W. J., 496.
Astronomy, problems of sidereal (Russell), 391.
ATKINSON, H. V., and LUSK, G. Production of Fat from Protein in the Dog, 246.
Atomic structure (Langmuir), 252.
Autumn Meeting of Academy, 87.
Award of Medals by the Academy, 492.

BAILEY, I. W. Phenomena of Cell Division in the Cambium of Arborescent Gymnosperms and their Cytological Significance, 283.
BARUS, C. Contact Lever, using Achromatic Displacement Fringes, 39.
——. Displacement of the Gravitating Needle in its Dependence on Atmospheric Temperatures, 547.
——. Interferential Contact Lever relating to the Elastics of Small Bodies, 44.
——. Self-Adjusting Interferometer in Relation to the Achromatic Fringes, 53.
——. Adjustment in Relation to the Fresnel Coefficient, 120.
——. Superposed or Duplicated Spectrum Fringes, 149.
——. Electrodynamometer using the Vibration Telescope, 211.
BARUS, C. Elongation due to Magnetization, 267.
——. Interferometry of Rapid Vibrations Chiefly in Relation to Telephone Currents, 331.
——. Pressure Variations of Specific Heat of Liquids, 340.
BATEMAN, H. Radiation from a Moving Magneton, 367.
Begonia Phyllomaniaca (Smith), 36.
BENEDICT, F. G., MILES, W. R. and JOHNSON, A. Temperature of Human Skin, 218.
BLODGETT, K., see Lemon, H. B., 289.
BRIDGES, C. B., see Sturtevant, A. H., 168.
BRIDGMAN, P. W. Comparison of Certain Electrical Properties of Ordinary and Uranium Lead, 351.
Buckwheat, variations in (Zinn), 506.
Bud variation (Stout), 130.

CALKINS, G. N. Restoration of Vitality through Conjugation, 95.
CAMPBELL, E. D. Studies of Constitution of Steel, 426.
CARMICHAEL, R. D. Conditions Necessary and Sufficient for the Existence of a Stieltjes Integral, 551.
Carotinoids, effect of absence of (Palmer), 582.
CASTLE, W. E. Is the Arrangement of the Genes in the Chromosome Linear?, 25.
——. Linkage System of Eight Sex-Linked Characters of Drosophila Virilis (Data of Metz), 32.
——. Piebald Rats and the Theory of Genes, 126.
——. Inheritance of Quantity and Quality of Milk Production in Dairy Cattle, 428.
——. Are Genes Linear or Non-Linear in Arrangement? 500.
Cell division in Gymnosperms (Bailey), 283.
Centenarians, age at death (Pearl), 83.
Charcoal, adsorption by (Lemon and Blodgett), 289; (Lemon), 291.
Chiton Tuberculatus (Crozier and Arey), 496.
Chromatin and cytoplasm, qualitative relations (Hegner), 19.
Chromosome, genes non-linear (Castle), 25, 32, 500.
Circles, cyclic systems (Eisenhart), 555.
CLARKE, F. W., and STEIGER, G. Inorganic Constituents of Lobster Shells, 6.

CLARKE, J. M. Falklandia, 102.
Clusters and nebulae (Russell), 392.
Cohesion, adhesion, etc. (Harkins), 562.
Color of stars (Seares), 232.
Conformal mapping (Gronwall), 22, 248.
Conjugation, restoration of vitality (Calkins) 95.
Contact lever (Barus), 39, 44.
CONTIERI, A. W., see MacInnes, D. A., 321.
Covariants of binary modular groups, (Glenn), 107.
CROZIER, W. J., and AREY, L. B. Ethology of Chiton Tuberculatus, 496.
CROZIER, W. J., see Arey, L. B., 498.

Dairy cattle, milk production (Castle), 428.
DAVENPORT, C. B., see Love, A. G., 58.
DETWILER, S, R, Effects of Transplanting Limbs upon the Formation of Nerve Plexuses and the Development of Peripheral Neurones, 324.
Diffusion through membranes (Loeb), 440.
Digestibility of Graham flour (Langworthy and Deuel), 514.
Discoidal structure of the lithosphere (Willis), 377.
Diseases of white and colored troops (Love and Davenport), 58.
Drosophila (Castle), 25, 32.
Drosophila, tumor in (Stark), 573.
DUANE, W. and SHIMIZU, T., X-Ray Absorption Wave Lengths of Lead Isotopes, 198.

EISENHART, L. P. Transformations of Cyclic Systems of Circles, 555.
Elastics of small bodies (Barus), 44.
Electrodynamometer (Barus), 211.
Electrolytes, effect on sinosis (Loeb), 440.
ELLERMAN, F., see Seares, F. H., 242.
Embryological basis of mortality (Pearl), 593.
EMCH, A. Certain Class of Rational Ruled Surfaces, 222.
Energy, molecular (Harkins), 539, 562.
Entropy and molecular association (Harkins), 539.
Equation of state of powder gases (Webster), 286.
Ethology of Chiton Tuberculatus (Crozier and Arey), 496.
Ethyl thioglycollate (Rouiller), 145.
Ethylene chlorhydrin (Gomberg), 424.

Falklandia (Clarke), 102.
Fat production from protein (Atkinson and Lusk), 246.
Fellowships in Physics and Chemistry (N. R. C.), 313.
FERGUSON, J. B. and MERWIN, H. E. The Ternary System CaO-MgO-SiO_2, 16.
FISHER, W., see Webster, A. G., 79.
Fistulae (Halsted), 76.
Flagellate affinities of trichonympha (Kofoid and Swezy), 9.
Flotation, principles of (Harkins), 569.
Flour, digestibility (Langworthy and Deuel), 514.
Fowls, growth and laying (Palmer), 582.
FRANKLIN, E. C. Metallic Salts of Pyrrol, Indol and Carbazol, 580.
Fréchet's class L (Moore), 206.
Fresnel coefficient (Barus), 120.
Fucus spores, phototropism (Hurd), 201.

Galactic system (Russell), 392.
Genes, arrangement in chromosome (Castle), 25, 32.
Genes, theory of (Castle), 126, 500.
Genes, theory of (Plough), 167; (Sturtevant *et al.*), 168.
GLENN, O. E. Covariants of Binary Modular Groups, 107.
GOMBERG, M. Ethylene Chlorhydrin, 424.
GOTTSCHALK, V. H., see Millikan, R. A., 591.
Grants from trust funds, 492.
Gravitating needle (Barus), 547.
GRONWALL, T. H. Theorem of Power Series, with an Application to Conformal Mapping, 22.
——. Twist in Conformed Mapping, 248.
Groups, binary modular (Glenn), 107.
Groups with two operators (Miller), 272.
Growth rate of Helianthus (Reed and Holland), 135.
Gun, pressures in (Webster and Thompson), 259.
Guns, projectiles from (Abbot), 386.
Gymnosperms, cell division (Bailey), 283.

HALL, E. H. Thermo-Electric Equation $P = TdV/dT$ Once More, 197.
HALSTED, W. S. Congenital Arterio-Venous and Lymphatico-Venous Fistulae, 76.
HARKINS, W. D. Adhesion between Mercury, Water, and Organic Substances, and the Fundamental Principles of Flotation, 569.
——. Change of Molecular Kinetic Energy into Molecular Potential Energy: the Entropy Principle and Molecular Association, 539.
——. Cohesion, Internal Pressure, Adhesion, Tensile Strength, Tensile Energy, Negative Surface Energy, and Molecular Attraction, 562.
——. Electromagnetic Hypothesis of the Kinetics of Heterogeneous Equilibrium, and of the Structure of Liquids, 152.
Harmonics, zonal spherical (Webster and Fisher), 79.
HECHT, S. Temperature and the Duration of a Biological Process, 146.
HEGNER, R. W. Quantitative Relations between Chromatin and Cytoplasm, 19.
Heine-Borel-Lebesgue theorem (Moore), 206
Helianthus, growth rate (Reed and Holland), 135.

Heterogeneous equilibrium (Harkins), 152.
HOLLAND, R. H., see Reed, H. S., 135.
Horns, theory of (Webster), 275.
HURD, A. M. Orienting Effects of Monochromatic Lights of Equal Intensities of Fucus Spores and Rhizoids, 201.
Hydrogen overvoltage (MacInnes and Adler), 160; (MacInnes and Contieri), 321.
Hydrogen, ozone form (Wendt), 518.
Hypersurfaces (Lefschetz), 296.

Indeterminate equations (Lehmer), 111.
Interferometry, see Barus, C.
International Astronomical Union, 193.
Ionization by α-rays (Millikan *et al.*), 591.
Isotopic lead (Duane and Shimizu), 198; (Bridgman), 351.

JOHNSON, A., see Benedict, F. G., 218.
JOY, A. H., see Adams, W. S., 239.

KELLY, M. J., see Millikan, R. A., 591.
Kinetics of liquids (Harkins), 152.
KOFOID, C. A. and SWEZY, O. Flagellate Affinities of Trichonympha, 9.

LANGMUIR, I. Structure of Atoms and the Octet Theory of Valence, 252.
LANGWORTHY, C. F. and DEUEL, H. J. Effect of Milling on the Digestibility of Graham Flour, 514.
Lead, electrical properties (Bridgman), 351.
Lead isotopes (Duane and Shimizu), 198.
LEFSCHETZ, S. Real Folds of Abelian Varieties, 103.
——. Real Hypersurfaces contained in Abelian Varieties, 296.
LEHMER, D. N. General Solution of the Indeterminate Equation: $Ax + By + Cz + \cdots = r$, 111.
LEMON, H. B. Variations, due to Heat Treatment, in the Rate of Absorption of Air by Cocoanut Shell Charcoal, 291.
LEMON, H. B. and BLODGETT, K. Relative Adsorption of Mixtures of Oxygen and Nitrogen in Cocoanut Shell Charcoal, 289.
LEUSCHNER, A. O. Perturbations and Tables of the Minor Planets, 67.
Liquids, specific heat (Barus), 340.
Liquids, structure of (Harkins), 152.
Lithosphere, structure of (Willis), 377.
Lobster shells (Clarke and Steiger), 6.
LOEB, J. Influence of Ions on the Electrification and Rate of Diffusion of Water through Membranes, 440.
LOVE, A. G. and DAVENPORT, C. B. Comparison of White and Colored Troops in respect to Incidence of Disease, 58.
LUSK, G., see Atkinson, H. V., 246.
Lymphatico-venous fistulae (Halsted), 76.

MACINNES, D. A. and ADLER, L. Hydrogen Overvoltage, 160.
MACINNES, D. A. and CONTIERI, A. W. Applications of Variation of Hydrogen Overvoltage with the Pressure, 321.
Magnetic field of Sun (Seares, Van Maanen and Ellerman), 242.
Magnetization and elongation (Barus), 267.
Magneton, radiation from (Bateman), 367.
Magnitudes in star clusters (Shapley), 344, 434.
Medals awarded by the Academy, 492.
MELTZER, S. J., and WOLLSTEIN, M. Influence of Degeneration of One Vagus Nerve upon the Development of Pneumonia, 493.
MENZIES, A. W. C. New Measurements of the Vapor Pressure of Mercury, 558.
Mercury, vapor pressures (Menzies), 558.
MERRILL, G. P. Meteorite Falls and Finds considered with reference to their varying basicity, 37.
MERWIN, H. E., see Ferguson, J. B., 16.
Meteorite falls and finds, (Merrill) 37.
MILES, W. R., see Benedict, F. G., 218.
Milk production in cattle (Castle), 428.
MILLER, G. A. Groups involving only two Operators which are Squares, 272.
MILLIKAN, R. A., GOTTSCHALK, V. A. and KELLY, M. J. Effect upon the Atom of the Passage of an Alpha Ray through it, 591.
Molecular energy (Harkins), 539, 562.
MOORE, R. L. Most General Class L of Fréchet in which the Heine-Borel-Lebesgue Theorem holds true, 206.
MORGAN, T. H., see Sturtevant, A. H., 168.
Moropus (Osborn), 250.
Mortality, embryological basis (Pearl), 593.

NATIONAL ACADEMY OF SCIENCES. Report of the Autumn Meeting, 87.
——. Report of the Annual Meeting, 479.
——. Award of Medals, 492.
——. Research Grants from the Trust Funds of the Academy, 492.
NATIONAL RESEARCH COUNCIL. Meetings of Executive Board, 50, 91, 299, 360, 462, 532, 603.
——. Meetings of the Executive Board with Council of the National Academy of Sciences, 180, 354, 452, 523.
——. Meetings of Interim Committee, 447, 459, 469, 528, 599.
——. Meetings of International Research Council and of Affiliated Organizations at Brussels, beginning July 18, 1919, 473.
——. National Research Fellowships in Physics and Chemistry, 313.
——, Organization of, 184.
——, Organization of Division of Physical Sciences, 316.
NELSON, N. C. Archaeology of the Southwest: A Preliminary Report, 114.
Nerve plexuses and peripheral neurones (Detwiler), 324.
Nervous organization of a Nudibranch (Crozier and Arey), 498.
NICHOLSON, S. B., see Shapley, H., 417.

Nudibranch, nervous organization (Arey and Crozier), 498.

Operators in group theory (Miller), 272.
OSBORN, H. F. Seventeen Skeletons of Moropus; Probable Habits of this Animal, 250.
——. Palaeomastodon, the Ancestor of the Long-Jawed Mastodons only, 265.
Osmosis (Loeb), 440.

Palaeomastodon (Osborn), 265.
Paleogeography (Ruedemann), 1.
PALMER, L. S. Growth and Reproduction in Fowls in the Absence of Carotinoids and the Physiological Relations of Yellow Pigmentation to Egg Laying, 582.
Parallaxes of brighter stars (Adams and Strömberg), 228.
PEARL, R. Mean Age at Death of Centenarians, 83.
——. The Embryological Basis of Human Mortality, 593.
PFUND, A. H. A Vapor-free Vacuum Seal, 221.
PERRINE, C. D. Distribution of the Aphelia of the Secondary Bodies of the Solar System, 371.
Phone and phonometer (Webster), 173.
Phonograph, theory of (Webster), 275.
Phototropism, of Fucus spores and rhizoids, (Hurd), 201.
Planets, perturbations of (Leuschner), 67.
PLOUGH, H. H. Linear Arrangement of Genes and Double Crossing Over, 167.
Pneumonia and the Vagus Nerve (Meltzer and Wollstein), 493.
Powder gases (Webster), 286.
Power series (Gronwall), 22.
Pre-Cambrian paleogeography (Ruedemann), 1.
Pressure in guns (Webster), 259.
Projectiles from guns, rotating (Abbot), 386.
Projectiles, speed of (Abbot), 388.
Proliferation in Begonia Phyllomaniaca (Smith), 36.
Protein producing fat (Atkinson and Lusk), 246.
Perturbations of the Watson Planets (Leuschner), 67.
Pyrrol, etc. (Franklin), 580.

Radiation from magneton (Bateman), 367.
Radiation, solar (Abbot), 383.
Radiationless orbits (Wilson), 588.
Rats, theory of genes (Castle), 126.
REED, H. S. and HOLLAND, R. H. Growth Rate of an Annual Plant Helianthus, 135.
Report of the annual meeting of the Academy, 479.
Report of the autumn meeting, 87.
RESEARCH COUNCIL, see National Research Council.
Research grants from the trust funds of the Academy, 492.
Rhizoids, phototropism of (Hurd), 201.
ROUILLER, C. A. Metallic Derivatives of Ethyl Thioglycollate, 145.
RUEDEMANN, R. Some Fundamentals of Pre-Cambrian Paleogeography, 1.
RUSSELL, H. N. Problems of Sidereal Astronomy, 391.

Sand, angle of repose (Webster), 263.
SEARES, F. H. Relation of Color to Intrinsic Luminosity in Stars of the same Spectral Type, 232.
SEARES, F. H., VAN MAANEN, A. and ELLERMANN, F. Deviations of Sun's General Magnetic Field from that of a uniformly Magnetized Sphere, 242.
SHAPLEY, H. Magnitudes in Star Clusters, IX, the Distances and Distribution of Seventy Open Clusters, 344.
——. Magnitudes in Star Clusters, X. Spectral Type B and the Local Stellar System, 434.
SHAPLEY H. and NICHOLSON, S. B. Spectral Lines of a Pulsating Star, 417.
SHIMIZU, T., see Duane, W., 198.
Sidereal Astronomy, problems of (Russell), 391.
Skin, temperature of (Benedict, Miles and Johnson), 218.
SMITH, ERWIN F. The Cause of Proliferation in Begonia Phyllomaniaca, 36.
Solar Constant of Radiation (Abbot), 383.
Solar system, aphelia in (Perrine), 371.
Southwest, archaeology of (Nelson), 114.
Spatial relations of genes (Sturtevant, Bridges and Morgan), 168.
Specific heat of liquids (Barus), 340.
Spectra of stars (Russell), 392.
Spectral lines of pulsating star (Shapley and Nicholson), 417.
Spectral type of stars (Seares), 232.
Spectrum, energy distribution (Webster), 163.
Spectrum fringes (Barus), 149.
Speed of projectiles (Abbot), 388.
Star clusters, magnitudes in (Shapley), 344, 434.
STARK, M. B. Benign Tumor that is Hereditary in Drosophila, 573.
Stars of high radial velocity (Adams and Joy), 239.
Star streaming (van Maanen), 225.
Steel, constitution of (Campbell), 426.
STEIGER, G., see Clarke, F. W., 6.
Stellar parallaxes (Adams and Strömberg), 228.
Stieltjes integral (Carmichael), 551.
STOUT, A. B. Bud Variation, 130.
Stream Motion, stellar (van Maanen), 225.
STRÖMBERG, G., see Adams, W. S., 228.
STURTEVANT, A. H., BRIDGES, C. B., and MORGAN T. H. Spatial Relations of Genes, 168.

Sun's magnetic field (Seares, van Maanen, and Ellerman), 242.
Surfaces, ruled (Emch), 222.
SWEZY, O., see Kofoid, C. A., 9.

Telephone currents and interferometry (Barus), 331.
Temperature effect in biology (Hecht), 146.
Temperature of human skin (Benedict, Miles and Johnson), 218.
Ternary system (Ferguson and Merwin), 16.
Thermo-electric equation (Hall), 197.
Thioglycollate (Rouiller), 145.
THOMPSON, L. T. E., see Webster, A. G., 259.
Transplanting limbs (Detwiler), 324.
Trichonympha (Kofoid and Swezy), 9.
Troops, white and colored (Love and Davenport), 58.
Tumors in Drosophila (Stark), 573.

Vacuum seal (Pfund), 521.
Vagus nerve and pneumonia (Meltzer and Wollstein), 493.
Valence, theory of (Langmuir), 252.
VAN MAANEN, A. Stream Motion afforded by Faint Stars near the Orion Nebula, 225.
——, see Seares, F. H., 242.
Vapor pressures of mercury (Menzies), 558.
Variation (Stout), 130.
Variation in buckwheat (Zinn), 506.
Varieties, abelian (Lefschetz), 103.
Velocities of stars (Adams and Joy), 239.
Vibration telescope (Barus), 211.
Vitality, restoration through conjugation (Calkins), 95.

WEBSTER, A. G. Complete Apparatus for Absolute Acoustical Measurements, 173.
——. Angle of Repose of Wet Sand, 263.
——. Acoustical Impedance and the Theory of Horns and of the Phonograph, 275.
——. Possible form of Equation of State of Powder Gases, 286.
WEBSTER, A. G., and FISHER, W. Tables of the Zonal Spherical Harmonic of the Second Kind $Q_1(z)$ and $Q_1(z)$, 79.
WEBSTER, A. G. and THOMPSON, L. T. E. New Instrument for Measuring Pressures in a Gun, 259.
WEBSTER, D. L. Approximate Law of Energy Distribution in the General X-Ray Spectrum, 163.
WENDT, G. L. Ozone Form of Hydrogen, 518.
WILLIS, B. Discoidal Structure of the Lithosphere, 377.
WILSON, EDWIN B. Radiationless Orbits, 588.
WOLLSTEIN, M., see Meltzer, S. J., 493.

X-ray absorption (Duane and Shimizu), 198.
X-ray spectrum (Webster), 163.

ZINN, J. On Variation in Tartary Buckwheat, Fagopyrum Tataricum (L.) Gaertn., 506.
Zonal spherical harmonics (Webster and Fisher), 79.